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# DOE STANDARD

## GUIDE OF GOOD PRACTICES FOR OCCUPATIONAL RADIOLOGICAL PROTECTION IN PLUTONIUM FACILITIES



**U.S. Department of Energy**  
**Washington, D.C. 20585**

**AREA SAFT**

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*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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**Foreword**

This Technical Standard does not contain any new requirements. Its purpose is to provide guides to good practice, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. The technical rationale is given to allow U.S. Department of Energy (DOE) health physicists to adapt the recommendations to similar situations throughout the DOE complex. The Standard provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, Occupational Radiation Protection. The Standard also supplements the DOE Implementation Guides, DOE Orders, and DOE's Radiological Control Manual and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling.

This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

Copies of electronic files of this Technical Standard may be obtained from the DOE Office of Worker Protection Programs and Hazards Management (WPPHM) Home Page Internet site (<http://tis-nt.eh.doe.gov/wpphm/regs/regs.html>). Copies of the Standard are also available from the DOE Technical Standards Program Internet site (<http://apollo.osti.gov/html/techstds/techstds.html>).

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U.S. DEPARTMENT OF ENERGY

GUIDE OF  
GOOD PRACTICES FOR OCCUPATIONAL  
RADIOLOGICAL PROTECTION IN  
PLUTONIUM FACILITIES

**Coordinated and Conducted  
for  
Office of Environment, Safety & Health  
U.S. Department of Energy**

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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**Table of Contents**

1.0	INTRODUCTION .....	1-1
1.1	PURPOSE AND APPLICABILITY .....	1-1
1.2	DEFINITIONS .....	1-2
1.3	DISCUSSION .....	1-2
2.0	MANUFACTURE, PROPERTIES, AND HAZARDS .....	2-1
2.1	MANUFACTURE OF PLUTONIUM .....	2-1
	2.1.1 Future Sources of Plutonium .....	2-4
	2.1.2 Laser Isotope Separation Process .....	2-5
2.2	NUCLEAR PROPERTIES .....	2-6
	2.2.1 Decay Schemes .....	2-7
	2.2.2 Neutron Yields and Spectra .....	2-8
2.3	PHYSICAL AND CHEMICAL PROPERTIES .....	2-12
	2.3.1 Plutonium Metal .....	2-13
	2.3.2 Plutonium Alloys .....	2-14
	2.3.3 Plutonium Compounds .....	2-15
	2.3.3.1 Plutonium Nitrate, Oxalate, Peroxide, and Fluorides .....	2-16
	2.3.3.2 Plutonium Dioxide .....	2-17
	2.3.3.3 Plutonium Hydride .....	2-18
	2.3.3.4 Plutonium Sulfates .....	2-18
	2.3.3.5 Plutonium Chlorides .....	2-19
	2.3.3.6 Plutonium Fuels .....	2-19
2.4	RADIOLOGICAL EFFECTS ON HUMANS .....	2-20
	2.4.1 Modes of Entry into the Body .....	2-20
	2.4.2 Distribution Within the Body .....	2-21
	2.4.3 Transfer to the Fetus .....	2-23
2.5	RADIATION EFFECTS ON MATERIALS .....	2-24
	2.5.1 Self-Heating .....	2-25
	2.5.2 Radiolysis .....	2-26
	2.5.2.1 Hydrogen Production .....	2-27
	2.5.2.2 Redox Reactions .....	2-28
	2.5.2.3 Miscellaneous Radiolytic Reactions .....	2-28
	2.5.2.4 Helium Retention and Release .....	2-29
2.6	OCCUPATIONAL HAZARDS .....	2-29
	2.6.1 Chemical Versus Radiological Hazards .....	2-30
	2.6.2 Associated Chemical Hazards .....	2-31
	2.6.3 Hazards Created by Oxidation and Pyrophoricity .....	2-31
	2.6.3.1 Oxidation of Plutonium .....	2-34
	2.6.3.2 Ignition Temperatures and Pyrophoricity of Plutonium, Its Alloys, and Its Compounds .....	2-35
	2.6.3.3 Aerolization of Plutonium .....	2-36
2.7	STORAGE AND CONTAINMENT .....	2-36
3.0	RADIATION PROTECTION .....	3-1
3.1	REGULATION AND STANDARDS .....	3-2
3.2	RADIATION PROTECTION PROGRAMS .....	3-2
	3.2.1 Quality Assurance .....	3-2



***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

---

	3.2.2	Administrative Controls	3-3
	3.2.2.1	Radiation Protection Procedures	3-3
	3.2.2.2	Radiological Work Permits	3-4
	3.2.2.3	Radiological Surveys and Data Trending	3-5
	3.2.2.4	Facility Posting and Labeling	3-10
	3.2.2.5	Unposted Areas	3-11
	3.2.3	Visitors	3-11
	3.2.4	Visits by Regulatory Personnel	3-13
	3.2.5	Onsite Packaging and Transportation	3-13
3.3		RADIOLOGICAL CONTROL ORGANIZATIONS	3-15
	3.3.1	Management Commitment	3-16
	3.3.2	Radiological Control Organization Independence and Reporting Level	3-16
	3.3.3	Adequacy of Personnel and Equipment	3-17
	3.3.4	Assignment of ALARA Responsibility and Authority	3-17
3.4		STAFFING AND STAFF QUALIFICATIONS	3-17
	3.4.1	Professional Staffing and Qualifications	3-18
	3.4.2	Technician Staffing and Qualifications	3-18
	3.4.3	Staffing Levels	3-19
3.5		INSTRUMENTATION CONSIDERATIONS	3-19
	3.5.1	Types of Instruments and Measurements	3-19
	3.5.2	General Performance Criteria for Instruments	3-21
	3.5.2.1	Portable Survey Instruments	3-21
	3.5.2.2	Performance Criteria for Fixed Monitoring Instruments	3-24
	3.5.2.3	Performance Criteria for Emergency Instrumentation	3-26
	3.5.3	Instrument Calibrations and Testing	3-28
3.6		RADIATION SAFETY TRAINING	3-30
	3.6.1	Radiological Worker Training	3-31
	3.6.2	Radiological Control Technician Training	3-31
	3.6.3	Training for Other Facility Personnel	3-31
	3.6.4	General Public Education	3-31
	3.6.5	Training Qualifications	3-32
	3.6.6	Health Physicist Training Involvement	3-32
3.7		RADIOLOGICAL RECORDS	3-32
3.8		ALARA AND OPTIMIZATION	3-33
	3.8.1	Current Status of ALARA Programs	3-34
	3.8.2	Achievement of Goals	3-34
	3.8.3	Quality Assurance	3-34
	3.8.4	Technical Aspects	3-35
	3.8.5	Attributes of Effective Review and Audit	3-35
3.9		CONDUCT OF OPERATIONS	3-39
	3.9.1	Radiation Work Procedures	3-41
	3.9.2	Posting and Labeling	3-41
	3.9.3	Calibration of Instruments	3-42
	3.9.4	Audits	3-42
	3.9.5	Decommissioning of Weapons and Weapon Facilities	3-43
4.0		CONTAMINATION CONTROL	4-1
4.1		AIR CONTAMINATION CONTROL	4-1
	4.1.1	Internal Versus External Dose Philosophy	4-2

***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

---

	4.1.2	Purpose of Air Monitoring .....	4-2
	4.1.3	Regulations and Limits .....	4-3
	4.1.4	Uncertainties and Limitations .....	4-3
	4.1.5	Samples and Instrumentation .....	4-4
	4.1.6	Sample Analysis .....	4-6
	4.1.7	Monitoring Strategies and Protocols .....	4-6
4.2		SURFACE CONTAMINATION CONTROL .....	4-7
	4.2.1	Plant Surfaces .....	4-7
	4.2.1.1	Housekeeping .....	4-9
	4.2.1.2	Vacuuming .....	4-11
	4.2.2	Reporting and Documenting Contamination Levels .....	4-11
	4.2.3	Characteristics of Plutonium Contamination .....	4-13
	4.2.4	Monitoring .....	4-13
	4.2.4.1	Contamination Surveys of the Workplace .....	4-15
	4.2.4.2	Release Surveys .....	4-16
	4.2.4.3	External Exposure Surveys .....	4-16
	4.2.4.4	Measurement and Survey Techniques .....	4-16
	4.2.5	Release Criteria .....	4-20
	4.2.6	Plutonium Contamination Detection .....	4-21
	4.2.7	ALARA Guideline .....	4-22
4.3		PERSONNEL CONTAMINATION CONTROL .....	4-23
	4.3.1	Monitoring Philosophy .....	4-23
	4.3.2	Monitoring Program .....	4-23
	4.3.3	Protective Clothing .....	4-24
	4.3.3.1	Requirements for Routine Operations .....	4-24
	4.3.3.2	Requirements for Special Maintenance .....	4-25
	4.3.4	Respiratory Protection .....	4-27
	4.3.5	ALARA Guidelines .....	4-28
	4.3.6	Release Criteria .....	4-28
4.4		Personnel Decontamination .....	4-28
5.0		INTERNAL DOSIMETRY .....	5-1
5.1		INTERNAL DOSE EVALUATION PROGRAM .....	5-2
	5.1.1	Performance Capabilities for Internal Exposure Monitoring .....	5-3
	5.1.2	Protection of the Embryo/Fetus, Minors, and Members of the Public .....	5-13
5.2		CHARACTERIZATION OF INTERNAL HAZARDS .....	5-13
5.3		SCOPE OF BIOASSAY PROGRAM .....	5-16
	5.3.1	Classification of Bioassay Measurements .....	5-17
	5.3.2	Monitoring Requirements and Selection of Employees .....	5-18
	5.3.3	Selection of Bioassay Monitoring Techniques .....	5-19
	5.3.3.1	In Vivo Counting .....	5-20
	5.3.3.2	In Vitro Analysis .....	5-22
5.4		ESTABLISHING BIOASSAY FREQUENCY .....	5-24
	5.4.1	Frequency Based on Program Sensitivity .....	5-25
	5.4.2	Frequency Based on Potential Risk of Intake .....	5-25
	5.4.3	Special Bioassay as Supplements to Routine Bioassay Programs ...	5-27
	5.4.4	Long-term Follow-up Bioassay Programs .....	5-27
5.5		ADMINISTRATION OF A BIOASSAY PROGRAM .....	5-28
	5.5.1	In Vivo Monitoring .....	5-29

***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

---

	5.5.2	Urine Sampling	5-30
	5.5.3	Fecal Sampling	5-33
5.6		MODELING THE BEHAVIOR OF PLUTONIUM IN AN IN THE BODY	5-34
	5.6.1	Respiratory Tract	5-34
	5.6.2	Gastrointestinal Tract	5-35
	5.6.3	Systemic Retention and Excretion of Plutonium	5-36
	5.6.4	Natural Plutonium Balance in an in Man	5-37
	5.6.5	Mother-to-Fetus Transfer	5-37
5.7		INTERPRETATION OF BIOASSAY RESULTS	5-38
	5.7.1	In an in Vivo Count Results	5-40
	5.7.2	Urine Sample Results	5-42
	5.7.3	Fecal Sample Results	5-43
	5.7.4	Use of Air Sample Data in an in Internal Dosimetry	5-44
5.8		DOSE ASSESSMENT	5-45
	5.8.1	Methods of Estimating Intake	5-47
	5.8.2	Alternate Methods of Intake Assessment	5-49
	5.8.3	Estimating Effective Dose Equivalent from Intakes of Plutonium	5-49
	5.8.4	Evaluating <sup>241</sup> Am Ingrowth In an in Vivo	5-50
5.9		INDICATOR AND ACTION LEVELS	5-53
5.10		RESPONSE TO SUSPECTED INTAKES	5-56
	5.10.1	Planning	5-58
	5.10.2	Medical Response Plan	5-58
	5.10.3	Responsibilities for Management of Internal Contamination	5-59
	5.10.4	Immediate Medical Care	5-60
	5.10.5	Contaminated Wounds	5-61
6.0		EXTERNAL DOSE CONTROL	6-1
6.1		DOSE LIMITS	6-2
	6.1.1	Limiting Quantities	6-2
	6.1.2	Operational Quantities	6-6
6.2		RADIATIONS IN PLUTONIUM FACILITIES	6-7
	6.2.1	Alpha and Beta Doses	6-7
	6.2.2	Gamma Doses	6-8
	6.2.2.1	Measured Gamma Dose Rates	6-10
	6.2.2.2	Calculated Photon Dose Rates	6-13
	6.2.3	Neutron Dose Equivalents	6-16
	6.2.3.1	Calculated Neutron Dose Equivalent Rates	6-18
	6.2.3.2	Neutron Emission Yields	6-19
	6.2.3.3	Quality Factors for Neutrons	6-23
6.3		RADIATION DETECTION AND EVALUATION	6-26
	6.3.1	Response of Portable Survey Instruments	6-26
	6.3.2	Personnel Dosimetry	6-28
	6.3.3	Extremity Dosimetry	6-32
	6.3.4	Criticality Accident Dosimetry	6-35
	6.3.5	Dose to Lens of Eye	6-40
	6.3.6	Spectrometry Measurements	6-40
	6.3.6.1	Multisphere Spectrometer System	6-41
	6.3.6.2	Tissue Equivalent Proportional Counter	6-43
	6.3.6.3	Liquid Scintillator Spectrometer	6-44
	6.3.6.4	Proton Recoil and Other Spectrometers	6-45

***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

---

6.4	EXTERNAL DOSE REDUCTION .....	6-46
6.4.1	Time .....	6-46
6.4.2	Distance .....	6-47
6.4.3	Shielding .....	6-47
6.4.3.1	Photon Shielding .....	6-48
6.4.3.2	Neutron Shielding .....	6-52
7.0	NUCLEAR CRITICALITY SAFETY .....	7-1
7.1	REGULATIONS AND STANDARDS .....	7-2
7.2	CRITICALITY CONTROL FACTORS .....	7-4
7.2.1	Technical Control Factors .....	7-4
7.2.2	Double Contingency .....	7-5
7.2.3	Administrative Control Factors .....	7-7
7.2.3.1	Nuclear Criticality Safety Program .....	7-7
7.2.3.2	Nuclear Criticality Safety Organization .....	7-8
7.2.3.3	Plans and Procedures .....	7-8
7.2.3.4	Nuclear Facility Safety Analysis .....	7-9
7.3	CRITICALITY ACCIDENT EXPERIENCE .....	7-10
7.3.1	Types of Criticality Accidents .....	7-10
7.3.2	Summary of Past Criticality Accidents .....	7-11
7.3.2.1	Los Alamos Accident - December 30, 1958 .....	7-11
7.3.2.2	Hanford-Recuplex Plant Accident - April 7, 1962 .....	7-12
7.4	CRITICALITY ALARMS AND NUCLEAR ACCIDENT DOSIMETRY .....	7-12
7.4.1	Criticality Alarm System .....	7-13
7.4.2	Nuclear Accident Dosimetry .....	7-15
7.4.2.1	Initial Screening Evaluation .....	7-16
7.4.2.2	Fixed and Personnel Nuclear Accident Dosimeters .....	7-17
7.4.2.3	Biological Indicators .....	7-20
7.5	RESPONSIBILITIES OF HEALTH PHYSICS STAFF .....	7-20
7.5.1	Routine Operations .....	7-20
7.5.2	Emergency Response Actions .....	7-21
7.5.3	Special Considerations During Decommissioning Activities .....	7-22
7.6	DEPARTMENT OF ENERGY PLUTONIUM VULNERABILITY ANALYSIS STUDY .....	7-22
8.0	WASTE MANAGEMENT .....	8-1
8.1	POTENTIALLY CONTAMINATED WASTES .....	8-1
8.1.1	Waste Types .....	8-1
8.1.2	Treatability Groups .....	8-2
8.1.3	Waste Disposal .....	8-5
8.2	AIRBORNE WASTE .....	8-7
8.2.1	Design Objectives .....	8-7
8.2.2	Operational Controls .....	8-9
8.2.3	Waste Treatments .....	8-9
8.2.4	Sampling and Monitoring .....	8-9
8.2.5	Disposal .....	8-10
8.3	SOLID WASTE .....	8-11
8.3.1	Design Objectives .....	8-11
8.3.2	Operational Controls .....	8-12
8.3.2.1	Waste Minimization .....	8-12

***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

---

	8.3.2.2	Waste Classification Control	8-15
	8.3.3	Waste Treatments	8-15
	8.3.4	Sampling and Monitoring	8-17
	8.3.5	Storage and Disposal	8-17
8.4		LIQUID WASTE	8-18
	8.4.1	Design Objectives	8-18
	8.4.2	Operational Controls	8-20
	8.4.3	Waste Treatments	8-20
	8.4.3.1	pH Adjustment	8-21
	8.4.3.2	Precipitation and Co-precipitation	8-21
	8.4.3.3	Liquid-Solid Separation Techniques	8-22
	8.4.3.4	Ion Exchange	8-22
	8.4.3.5	Distillation	8-24
	8.4.3.6	Purification by Reverse Osmosis	8-24
	8.4.3.7	Solidification	8-25
	8.4.3.8	Solvent Extraction	8-25
	8.4.3.9	Incineration	8-25
	8.4.4	Sampling and Monitoring	8-25
	8.4.5	Storage and Disposal	8-26
9.0		EMERGENCY MANAGEMENT	9-1
9.1		Emergency Management In DOE	9-1
	9.1.1	Key Emergency Management Principles	9-1
	9.1.2	Requirements Pertaining to All DOE Operations	9-3
9.2		Specific Guidance on Emergency Management For Plutonium Facilities	9-3
	9.2.1	Hazards Assessment	9-4
	9.2.2	Program Elements	9-8
10.0		DECONTAMINATION AND DECOMMISSIONING	10-1
10.1		REGULATIONS AND STANDARDS	10-1
	10.1.1	Other Regulations	10-3
	10.1.2	Residual Radioactivity Levels	10-4
10.2		DESIGN FEATURES	10-8
	10.2.1	Building Materials	10-8
	10.2.2	Ventilation Systems	10-9
	10.2.3	Piping Systems	10-10
	10.2.4	Soil-Contamination Considerations	10-10
	10.2.5	Other Features	10-10
10.3		DECONTAMINATION AND DECOMMISSIONING PROGRAM REQUIREMENTS	10-11
10.4		DECONTAMINATION AND DECOMMISSIONING TECHNIQUES	10-13
	10.4.1	Equipment and Surface Decontamination	10-13
10.5		DECONTAMINATION AND DECOMMISSIONING EXPERIENCE	10-14
11.0		REFERENCES	11-1
APPENDIX A			
		GLOSSARY	A-1

***Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***

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## APPENDIX B

PLUTONIUM IN DEPARTMENT OF ENERGY FACILITIES .....	B-1
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## APPENDIX C

FACILITY DESIGN .....	C-1
-----------------------	-----

**FIGURES**

2.1. Principal Modes of Plutonium Production by Neutron Irradiation of Uranium .....	2-3
2.2. Atom Ratio of $^{241}\text{Am}$ to $^{241}\text{Pu}$ ( $t=0$ ) Produced by the Beta Decay of $^{241}\text{Pu}$ as a Function of Time Since Chemical Separation .....	2-11
2.3. Neutron Energy Spectra of Plutonium-Beryllium and Plutonium-Boron Neutron Sources Compared with a Fission Source .....	2-12
2.4. Hazards in Low-Exposure Plutonium Handling .....	2-30
6.1. Absorbed Surface Dose Rate from Plutonium Dioxide as Measured with an Extrapolation Chamber .....	6-9
6.2. Energy Dependence of Various TLD-Albedo Dosimeters .....	6-31
6.3. Response of Electrochemically Etched CR-39 Used in Nuclear Track Dosimeters as a Function of Neutron Energy .....	6-31
6.4. Fixed Nuclear Accident Dosimeter Used at Hanford to Help Assess Doses from Criticality Accidents .....	6-39
6.5. Neutron Energy Spectra as Measured by the Multisphere Spectrometer at 50 cm from Plutonium Metal, $\text{PuO}_2$ , and $\text{PuF}_4$ Sources .....	6-45
6.6. Reduction in Photon Dose Rate with Various Shielding Materials at a Distance of 3 cm from a 100-gram Disk of Plutonium Oxide .....	6-51
6.7. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Tetrafluoride Sources .....	6-52
6.8. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Oxide Sources .....	6-53

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

## TABLES

2.1.	Isotopic Composition of Three Grades of Plutonium: Heat Source, Weapons, and Reactor	2-4
2.2.	Uses and Availabilities of Plutonium Isotopes	2-5
2.3.	Radioactive Decay Properties of Selected Isotopes and Decay Products, Excluding Spontaneous Fission	2-9
2.4.	Specific Activity Decay Heats of Selected Isotopes	2-10
2.5.	Allotropic Forms of Plutonium Metal	2-13
2.6.	Solubilities and Properties of Selected Compounds	2-16
2.7.	Common Biokinetic Models for Plutonium and Americium	2-23
2.8.	Potential Hazards or Damage to Materials from Exposure to Radiation	2-25
2.9.	Hazards of Chemicals Used in Processing Plutonium	2-33
2.10.	Storage Recommendations for Plutonium Metal and Dioxide	2-39
4.1	Surface Radioactivity Values, dpm/100 cm <sup>2</sup>	4-4
5.1.	Urine Bioassay Goals for <sup>239</sup> Pu	5-6
5.2.	Fecal Bioassay Goals for <sup>239</sup> Pu	5-8
5.3	In Vivo Lung Measurement Bioassay Goals for <sup>241</sup> Am as an Indicator of Aged Weapons-Grade Plutonium	5-10
5.4.	Example Plutonium Isotope Mixtures Immediately Post-Separation, wt%	5-15
5.5	Activity Composition with Age for Reference 6% and 12% <sup>240</sup> Pu Mixtures	5-16
5.6.	Intake Retention Fractions for <sup>239</sup> Pu	5-48
5.7.	Plutonium or <sup>241</sup> Am Indicator Levels for	5-54
5.8.	Plutonium or <sup>241</sup> Am Contamination Levels for Notification of Occupational Medicine Physician	5-54
6.1.	Effective Depth of Tissue for Various Organs	6-2
6.2.	Tissue Weighing Factors	6-4
6.3.	Radiation Dose Limits for DOE and DOE Contractors	6-5
6.4.	Isotopic Composition of the Plutonium Used in the Extremity Dosimetry Measurements	6-11
6.5.	Gamma Dose Rates Along an Arm Phantom in Contact with a Steel Can Containing 1 kg of Plutonium Dioxide in an Uncontaminated Glove Box	6-11
6.6.	Gamma Dose Rates Measured with an Arm Phantom Placed Inside Gloves Dusted with Plutonium Dioxide Powder	6-13
6.7.	Spontaneous Fission Neutron Yields	6-20
6.8.	Neutron Yields from Alpha-Neutron Reactions for oxides and Fluorides	6-23
6.9.	Neutron Yields for Trace Impurities in Plutonium and Uranium	6-27
6.10.	Performance Test Categories, Radiation Sources, and Test Ranges for the DOELAP and NVLAP Programs	6-34
6.11.	Photon Dose Rates at 2 Meters from Cylinders of Plutonium Containing 1 kg of Plutonium at 5 Years After Chemical Separation	6-50
6.12.	Isotopic Composition of Plutonium Sources at 5 Years After Chemical Separation of the Plutonium	6-51
7.1.	Subcritical, Single Parameter Limits for Plutonium Solutions and Metals	7-6
8.1.	Waste Types	8-3
8.2.	Treatability Groups	8-4
8.3	Interim Recommendations for Storage of Plutonium Metal and Plutonium Oxide at Department of Energy Facilities	8-27

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## **1.0 INTRODUCTION**

### **1.1 PURPOSE AND APPLICABILITY**

This Technical Standard (TS) does not contain any new requirements. Its purpose is to provide guides to good practice, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. The technical rationale is given to allow U.S. Department of Energy (DOE) health physicists to adapt the recommendations to similar situations throughout the DOE complex. Generally, DOE contractor health physicists will be responsible to implement radiation protection activities at DOE facilities and DOE health physicists will be responsible for oversight of those activities. This guidance is meant to be useful for both efforts. The TS provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, Occupational Radiation Protection (DOE, 1993c); hereinafter referred to as 10 CFR 835. The TS also supplements the DOE Implementation Guides (IGs), DOE Orders, and DOE's Radiological Control Manual (DOE, 1994l) and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling. This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

This TS replaces PNL-6534, Health Physics Manual of Good Practices for Plutonium Facilities (PNL, 1988) by providing more complete and current information and by emphasizing the situations that are typical of DOE's current plutonium operations; safe storage, decontamination, and decommissioning (environmental restoration); and weapons disassembly.

The technical information presented here represents the best technical information available from within the DOE complex. Except to the extent that the guidance presented here duplicates mandatory regulations or contract requirements, it is not binding or mandatory. Any DOE Orders, manuals or guides, referred to in this TS are not binding unless they have been incorporated into the applicable contract to assist in identifying applicable requirements "shall" statements are followed by a reference. Should and my statements are provided for consideration. However, judicious use of this TS, along with

the regulatory documents discussed above, will help assure a comprehensive and technically defensible radiological protection program.

Regulatory guidance and references are current as of September 1997.

## **1.2 DEFINITIONS**

A glossary is provided (see Appendix A) to ensure uniform understanding of words in this document. In all cases, the definitions given here are consistent with those used in the Implementation Guides.

## **1.3 DISCUSSION**

Chapters 2 through 10 provide technical information to assist in safely managing plutonium operations. The topics covered are those considered by representatives of many of DOE's plutonium facilities to be most beneficial: Manufacture, Properties and Hazards, Radiation Protection, Contamination Control, Internal Dosimetry, External Dose Control, Nuclear Criticality Safety, Waste Management, Emergency Management, and Decontamination and Decommissioning. Appendixes B and C summarize information from other documents that are useful for reference: Plutonium in Department of Energy Facilities, and Facility Design.

## 2.0 MANUFACTURE, PROPERTIES, AND HAZARDS

This chapter briefly describes the manufacture of plutonium and presents the nuclear, physical, chemical, and radiobiological properties of plutonium (and/or sources for these data) that form the basis for radiological and toxic control limits. The data and discussion are intended to provide a basis for understanding the changes in hazards as a function of such parameters as isotopic composition, age since chemical processing, physical form, and chemical form. Data are presented to facilitate the calculation of radiation effects, which occur from a variety of plutonium sources.

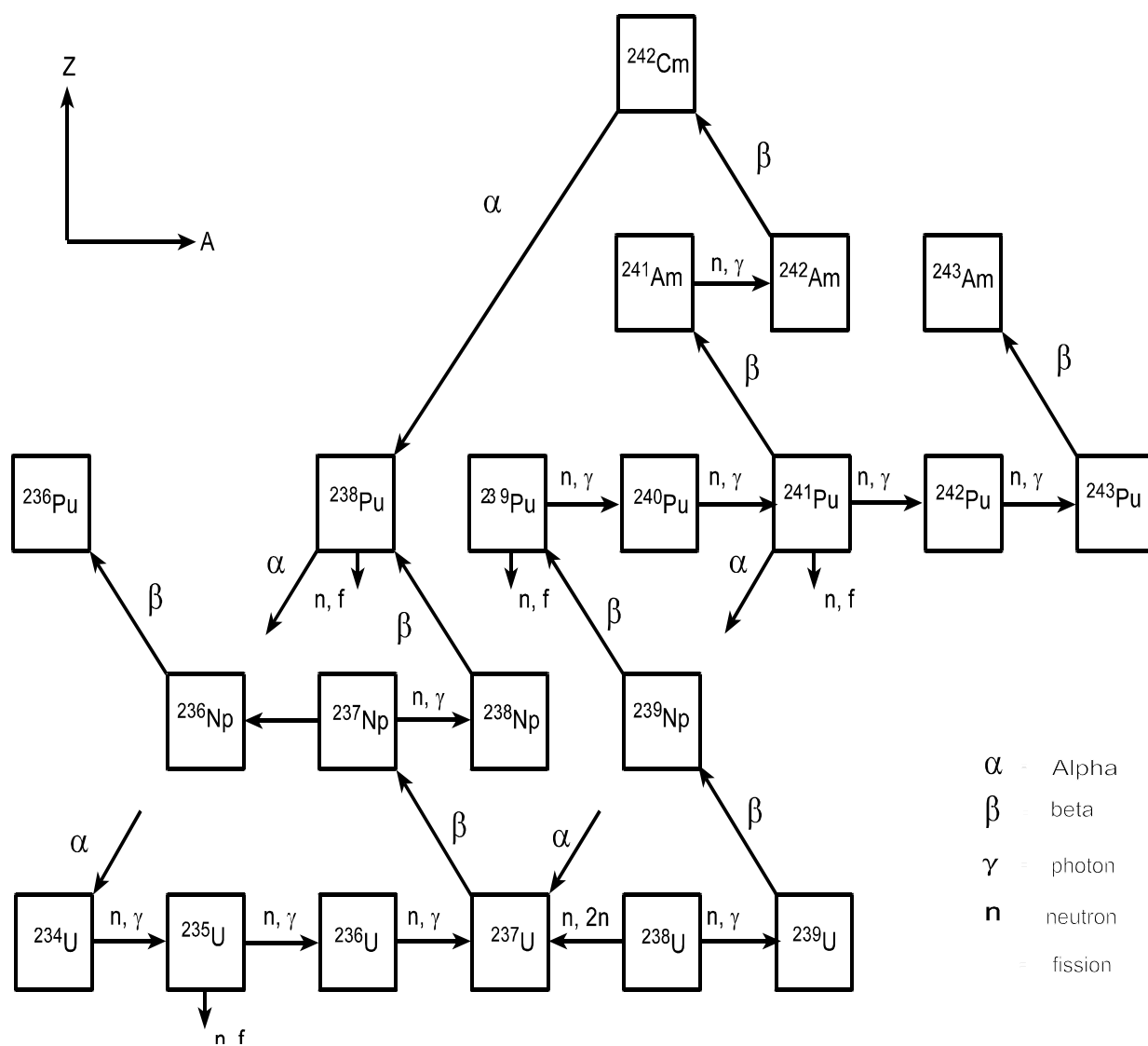
Plutonium is the first man-made element produced on an industrial scale. The special nuclear properties of  $^{239}\text{Pu}$  and  $^{238}\text{Pu}$  have led scientists to focus their efforts on these two isotopes. The fission cross-section of  $^{239}\text{Pu}$  makes it a useful energy source for atomic weapons and nuclear power reactors. The 87.7-year half-life of  $^{238}\text{Pu}$  makes it an excellent heat source for space applications. Unfortunately, the same nuclear properties of plutonium that make it attractive to science also make this element hazardous to human beings. All 15 plutonium isotopes are radioactive, with half-lives ranging from 26 minutes for  $^{235}\text{Pu}$  to  $7.6 \times 10^7$  years for  $^{244}\text{Pu}$ .

### 2.1 MANUFACTURE OF PLUTONIUM

Because of its high specific alpha activity and high decay heat,  $^{238}\text{Pu}$  has been used as an isotopic heat source for devices that generate thermoelectric power, such as the Space Nuclear Auxiliary Power (SNAP) systems used in lunar and deep space missions. Small amounts of  $^{238}\text{Pu}$  with low  $^{236}\text{Pu}$  content were used as a power source for medical prosthetic devices such as cardiac pacemakers and a prototype artificial heart, but lithium batteries have replaced these plutonium power sources.  $^{238}\text{Pu}$  containing a few parts per million of  $^{236}\text{Pu}$  is produced by irradiating  $^{237}\text{Np}$  with slow neutrons. It can also be produced by irradiating  $^{241}\text{Am}$  to form  $^{242}\text{Cm}$ , which quickly decays to  $^{238}\text{Pu}$ .

In the past, most plutonium in DOE facilities was produced for nuclear weapons and was composed of greater than 90 wt%  $^{239}\text{Pu}$  and about 6 to 8 wt%  $^{240}\text{Pu}$ . This material has been referred to as “weapons grade” or “low exposure” plutonium. It is produced on a large scale by irradiating  $^{238}\text{U}$  in moderated production reactors (see Figure 2.1). Plutonium has also been produced as a byproduct in the operation of research reactors, and commercial nuclear power plants. It is recovered and purified by solvent extraction and ion exchange processes. The resulting highly concentrated  $\text{Pu}(\text{NO}_3)_4$  product solution is converted to a nonhygroscopic  $\text{PuF}_4$  intermediate by one of several processes before being reduced to metal with calcium. Plutonium is also produced from the waste streams of

the conversion processes and scrap recovery operations, which include material from research and development efforts. Other processes for reduction to metal include direct reduction of the oxide and electrolytic reduction. Typical isotopic compositions of three common grades of plutonium are given in Table 2.1.

**Figure 2.1.** Principal Modes of Plutonium Production by Neutron Irradiation of Uranium

**Table 2.1.** Isotopic Composition of Three Grades of Plutonium: Heat Source, Weapons, and Reactor

Isotope	Heat Source	Weapons Grade	Reactor Grade
$^{238}\text{Pu}$	90.0	<0.05	1.5
$^{239}\text{Pu}$	9.1	93.6	58.1
$^{240}\text{Pu}$	0.6	6.0	24.1
$^{241}\text{Pu}$	0.03	0.4	11.4
$^{242}\text{Pu}$	<0.01	<0.05	4.9

Overviews of plutonium process chemistry at DOE's Hanford, Los Alamos, Rocky Flats, and Savannah River sites are given by Christensen et al. (1983), Baldwin and Navratil (1983), Coops et al. (1983), and Christensen and Mullins (1983). In each case, solutions for recovery, purification, and waste treatment operations are emphasized. Technology under consideration for incorporation in Weapons Complex 21 is described by Christensen (1992).

### 2.1.1 Future Sources of Plutonium

High-exposure plutonium, i.e., plutonium containing significant fractions of  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$ , is produced in power reactor fuels. Currently, this form of plutonium is in the irradiated fuel in spent-fuel storage basins and other sources resulting from development work performed to demonstrate plutonium fuel cycles. Because recycling of commercial reactor fuel is not anticipated, future supplies of plutonium will be primarily from DOE production facilities and from reprocessing of current material. In the more distant future, liquid metal fast breeder reactors (LMFBRs) may be a potential source of plutonium.

Special isotopes of reasonably high purity are also available, which can be useful to health physicists for calibration purposes. These isotopes and their sources are listed in Table 2.2.

New sources of plutonium include the return of atomic weapon components and plutonium recovered from decontamination and decommissioning (D&D) operations. Foreign plutonium from states of the former Soviet Union may become an additional source. Their weapons-grade plutonium is believed to contain 5%  $^{240}\text{Pu}$ . Americium is not periodically removed from their stockpile material.

### 2.1.2 Laser Isotope Separation Process

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

Several new technologies are being considered to provide more highly purified plutonium isotopes for various purposes. One of these processes, laser isotope separation (LIS), has the potential to purify  $^{239}\text{Pu}$  from almost any source of plutonium. The LIS process produces a product enriched in  $^{239}\text{Pu}$  and a byproduct that contains the remaining plutonium isotopes. It is conceivable that the byproduct stream could be further purified to produce a specific plutonium isotope, such as  $^{238}\text{Pu}$  used for isotopic heat sources.

**Table 2.2.** Uses and Availabilities of Plutonium Isotopes

Isotope	Uses	Availability
$^{236}\text{Pu}$ , $^{237}\text{Pu}$	Popular environmental and biological chemical tracers.	Both available in microcurie quantities. <sup>(a)</sup>
$^{238}\text{Pu}$	Small thermal and electric-power generators.	Available in various isotopic enrichments, ranging from 78% to 99+%. <sup>(a)</sup>
$^{239}\text{Pu}$	Nuclear weapons and as a fast reactor fuel. Also, frequently used in chemical research where production-grade material of mixed isotopic content is suitable.	Available enrichments range from 97% to 99.99+%. <sup>(a)</sup>
$^{240}\text{Pu}$	Principally in flux monitors for fast reactors.	Available enrichments range from 93% to 99+%. <sup>(a)</sup>
$^{241}\text{Pu}$	The parent from which high-assay $^{241}\text{Am}$ can be isolated for industrial purposes.	Samples available in enrichments of 93%. <sup>(a)</sup>
$^{242}\text{Pu}$	For study of the physical properties of plutonium; also as a mass spectroscopy tracer and standard.	Samples available in enrichments ranging from 95% to 99.9+%; enrichments of production-grade material range from 85% to 95%. <sup>(a)</sup>
$^{244}\text{Pu}$	Currently, the only isotope available as a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM). <sup>(b)</sup>	Can be obtained from DOE's New Brunswick Laboratory.

(a) Available in small quantities from the Oak Ridge National Laboratory (ORNL): ORNL Isotopes Sales Office, Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, Tennessee 37830.

(b) A second NIST/SRM (a 1:1 mixture of  $^{239}\text{Pu}$  and  $^{242}\text{Pu}$ ) is being prepared, and a third ( $^{239}\text{Pu}$ ) is planned for the future.

The LIS process has many benefits. It can significantly reduce external radiation exposure to both neutron and gamma radiations for the product enriched in  $^{239}\text{Pu}$ . (Potential exposure problems from the byproduct stream are discussed later in this section.) The International Commission on Radiological Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP) have recommended increasing quality factors to a value of 20 for fast neutrons (ICRP, 1985; NCRP, 1987a). Thus, it may be

desirable to reduce neutron exposures. Neutrons arise primarily from even-numbered plutonium isotopes (mostly  $^{238}\text{Pu}$  and  $^{240}\text{Pu}$ ) as a result of spontaneous fission and alpha-neutron reactions with low-atomic-number impurities in the plutonium. The  $^{239}\text{Pu}$ -enriched product of LIS will have reduced concentration of these isotopes, resulting in lower intrinsic neutron exposures. The LIS process can also result in significant reductions in gamma-ray exposures for the product enriched in  $^{239}\text{Pu}$ . Much of the whole-body and most of the extremity exposure is a result of surface contamination on the gloves and the interior of the glovebox. The  $^{241}\text{Am}$  decay product, which results from the beta decay of  $^{241}\text{Pu}$ , is a major contributor. Thus, the reduction of  $^{241}\text{Pu}$  can significantly reduce exposures to hands and arms, as well as reduce the radiation streaming through glove ports in shielded gloveboxes.

## 2.2 NUCLEAR PROPERTIES

Of the 15 plutonium isotopes, the two that have proven most useful are masses 239 and 238. Plutonium-239 is fissile, i.e., atoms of plutonium split upon exposure to thermal or fast neutrons. Chemical reactions can release a few electron volts of energy per atom; however, when a plutonium nucleus splits, it releases about 200 MeV of energy and two or three neutrons. This release of energy makes  $^{239}\text{Pu}$  useful for nuclear weapons and reactor fuel. In fact, in light water reactors (LWRs) much of the power originates from the fission of  $^{239}\text{Pu}$ , which is produced by neutron capture in  $^{238}\text{U}$ . Because of its higher specific activity,  $^{238}\text{Pu}$  is used as long-lived heat sources for powering planetary space missions where adequate solar energy is not available.

As mentioned before, all plutonium isotopes are radioactive. Isotopes with even mass numbers (except mass number 246) are primarily alpha emitters. Isotopes of mass numbers 232, 233, 234, 235, and 237 also decay by electron capture; isotopes of mass numbers 241, 243, 245, and 246 decay by beta emission. Many of the alpha-emitting isotopes, such as  $^{238}\text{Pu}$  and  $^{240}\text{Pu}$ , also fission spontaneously and emit neutrons. All of the particle emissions are accompanied by X-ray and gamma-ray emissions over a wide range of energies.

A review of the nuclear properties of plutonium (e.g., cross-sections, nuclear levels, half-lives, and fission yields) can be found in Volume 1 of the Plutonium Handbook: A Guide to the Technology (Wick, 1967) and in American National Standards Institute (ANSI) Standard N317, Performance



Criteria for Instrumentation Used for In-Plant Plutonium Monitoring (ANSI, 1980a). Plutonium decay schemes, neutron yields, and neutron energy spectra are described in the following sections.

### **2.2.1 Decay Schemes**

The decay modes of some important plutonium and other isotopes and decay products are shown in Table 2.3. For brevity, only the most abundant radiations have been included in the table; more detailed information can be found in papers by Gunnink and Morrow (1967) and Klein (1971), in ICRP Publication 38 (ICRP, 1983), and from the National Nuclear Data Center. Most of the isotopes are strong alpha-emitters, making alpha heating a problem for the storage and handling of large amounts of plutonium. The specific activities and decay heats for selected isotopes and decay products are given in Table 2.4. Kilogram quantities of  $^{239}\text{Pu}$  or gram quantities of  $^{238}\text{Pu}$  can generate enough heat to melt plastic bags. Sources of  $^{238}\text{Pu}$  must be handled with insulated gloves, and special precautions must be taken to ensure a good thermal heat sink during shipping and storage. (See also Section 2.5.1, “Self-Heating.”)

The plutonium isotopes emit relatively few high-energy gamma rays, so even kilogram quantities can be processed without serious gamma-exposure problems. Because of the high density of plutonium, many gamma rays are self-absorbed. In some instances, the decay products may become significant in radiation protection and metallurgy. For instance, the isotope  $^{236}\text{Pu}$  often constitutes less than 1% of plutonium and is often ignored in dose calculations. However, if the plutonium is shielded by greater than 1 cm of lead or steel, the decay products of  $^{236}\text{Pu}$  may be the largest contributors to exposure. The decay product  $^{208}\text{Tl}$  emits a highly penetrating gamma ray with an energy of 2.615 MeV. In plutonium that contains a few weight percent  $^{241}\text{Pu}$ , the  $^{241}\text{Am}$  decay product is important because it emits a large number of 60-keV photons, which can be a significant source of exposure to the hands and forearms when handling plutonium in gloveboxes. (See Section 6.3.3 for more information.) Also,  $^{241}\text{Am}$  can contribute to neutron dose. Americium-241 contributes to increased alpha emission which affects the neutron dose as well as radiolysis and helium retention and release. Because of its importance to radiation exposure, the fractional amount of  $^{241}\text{Am}$  produced by beta decay from  $^{241}\text{Pu}$  is given as a function of time since chemical separation (see Figure 2.2).

### **2.2.2 Neutron Yields and Spectra**

Plutonium and plutonium compounds also emit neutrons from spontaneous fission and from alpha-neutron reactions with light elements. The spontaneous fission half-life and the neutron yields from spontaneous fission and alpha-neutron reactions for plutonium metal and plutonium compounds are provided in Section 6.0 of this TS. The approximate neutron yield from a substance with a known isotopic composition can be determined by adding the contributions from each component. This procedure and its limitations are described in detail in Section 6.0, which also discusses neutron dose equivalent rates.

**Table 2.3.** Radioactive Decay Properties of Selected Isotopes and Decay Products, Excluding Spontaneous Fission<sup>(a)</sup>

Isotope	Half-Life	Mode of Decay Particle	Energy, MeV	Yield,d%	X-ray <sup>(b)</sup> Energy, MeV	Yield, %	Gamma Ray Energy, MeV	Yield, %
<sup>236</sup> Pu	2.851 y	α	5.77	69.3	L's 0.011-0.021	13 <sup>©</sup>	0.0476	6.6 x 10 <sup>-2</sup>
		α	5.72	30.6			0.109	1.2 x 10 <sup>-2</sup>
<sup>238</sup> Pu	87.7 y	α	5.50	71.0	L's 0.011-0.021	10.5 <sup>©</sup>	0.0425	3.95 x 10 <sup>-2</sup>
		α	5.46	28.8			0.0999	7.35 x 10 <sup>-3</sup>
<sup>239</sup> Pu	2.41 x 10 <sup>4</sup> y	α	5.157	73.1	L's 0.0116-0.0215	5.0 <sup>©</sup>	0.099	1.22 x 10 <sup>-3</sup>
		α	5.144	15.0			0.129	6.41 x 10 <sup>-3</sup>
		α	5.106	11.8			0.375	1.55 x 10 <sup>-3</sup>
							0.414	1.46 x 10 <sup>-3</sup>
<sup>240</sup> Pu	6564 y	α	5.168	72.8	L's 0.0115-0.0215	10.8 <sup>©</sup>	0.0452	4.50 x 10 <sup>-2</sup>
		α	5.124	27.1			0.104	7.08 x 10 <sup>-3</sup>
<sup>241</sup> Pu	14.35 y	β	0.0052 <sup>(d)</sup>	100.00	—	—	0.077	2.20 x 10 <sup>-5</sup>
		α	4.896	2.04 x 10 <sup>-3</sup>			0.1037	1.01 x 10 <sup>-4</sup>
							0.114	6.0 x 10 <sup>-6</sup>
							0.149	1.9 x 10 <sup>-4</sup>
							0.160	6.71 x 10 <sup>-5</sup>
<sup>242</sup> Pu	3.73 x 10 <sup>5</sup> y	α	4.901	77.5	L's 0.0116-0.0215	9.1 <sup>©</sup>	0.0449	3.6 x 10 <sup>-2</sup>
		α	4.857	22.4			0.104	7.8 x 10 <sup>-3</sup>
<sup>241</sup> Am	432.2 y	α	5.486	85.2	L's 0.0119-0.0222	42 <sup>©</sup>	0.0263	2.4
		α	5.443	12.8			0.0332	1.2 x 10 <sup>-1</sup>
		α	5.388	1.4			0.0595	35.7
<sup>237</sup> U	6.75 d	β	0.039 <sup>(e)</sup>	0.8	L's 0.0119-0.0206	70 <sup>©</sup>	0.0263	2.43
		β	0.050 <sup>(e)</sup>	3.4			0.0595	34.5
		β	0.065 <sup>(e)</sup>	51			0.0648	1.28
		β	0.069 <sup>(e)</sup>	42	K's 0.097-0.114	53	0.165	1.85
							0.208	21.1
							0.268	7.1 x 10 <sup>-1</sup>
							0.332	1.2
							0.335	9.5 x 10 <sup>-2</sup>
							0.369	4.0 x 10 <sup>-2</sup>
							0.371	1.1 x 10 <sup>-1</sup>

(a) Data from Dunford and Burrows (1993).

(b) L's = L X-rays; K's = K X-rays.

(c) Total for all X-rays. The value represents an average obtained from data at Pacific Northwest Laboratory, Lawrence Berkeley Laboratory, and Lawrence Livermore Laboratory.

(d) Average beta energy given. The maximum beta average for <sup>241</sup>Pu is 0.0208 MeV.(e) Average beta energy. The maximum beta energy for <sup>237</sup>U is 0.248 MeV.

**Table 2.4.** Specific Activity Decay Heats of Selected Isotopes<sup>(a)</sup>

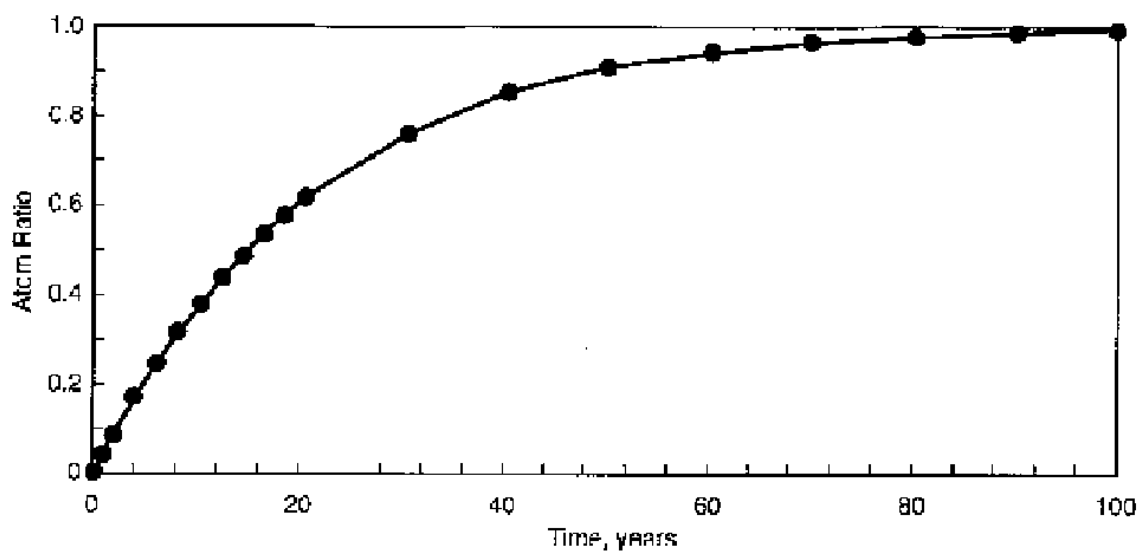
Isotope	Half-Life, y	Specific Activity, Ci/G		Average Particle Energy per		Decay Heat, W/g <sup>(b)</sup>
				Disintegration, MeV <sup>(b)</sup>		
<sup>236</sup> Pu	2.851	α	534	α	5.75	18.2
<sup>238</sup> Pu	87.7	α	17.1	α	5.49	0.567
<sup>239</sup> Pu	2.407 x 10 <sup>4</sup>	α	6.22 x 10 <sup>-2</sup>	α	5.14	1.93 x 10 <sup>-3</sup>
<sup>240</sup> Pu	6564	α	0.229	α	5.16	7.13 x 10 <sup>-3</sup>
<sup>241</sup> Pu	14.35	α	2.52 x 10 <sup>-3</sup>	α+β	5.27 x 10 <sup>-3</sup>	3.29 x 10 <sup>-3</sup>
		β	103			
<sup>242</sup> Pu	3.733 x 10 <sup>5</sup>	α	3.93 x 10 <sup>-3</sup>	α	4.90	1.16 x 10 <sup>-4</sup>
<sup>232</sup> U	72.0	α	21.5	α	5.31	0.690
<sup>233</sup> U	1.59 x 10 <sup>5</sup>	α	9.75 x 10 <sup>-3</sup>	α	4.72	2.84 x 10 <sup>-4</sup>
<sup>234</sup> U	2.45 x 10 <sup>5</sup>	α	6.29 x 10 <sup>-3</sup>	α	4.76	1.81 x 10 <sup>-4</sup>
<sup>235</sup> U	7.04 x 10 <sup>7</sup>	α	2.17 x 10 <sup>-6</sup>	α	4.24	6.02 x 10 <sup>-6</sup>
<sup>236</sup> U	2.34 x 10 <sup>7</sup>	α	6.5 x 10 <sup>-5</sup>	α	4.48	1.77 x 10 <sup>-6</sup>
<sup>238</sup> U	4.47 x 10 <sup>9</sup>	α	3.38 x 10 <sup>-7</sup>	α	4.18	8.58 x 10 <sup>-9</sup>
<sup>237</sup> Np	2.14 x 10 <sup>6</sup>	α	7.08 x 10 <sup>-4</sup>	α	4.76	2.08 x 10 <sup>-5</sup>
<sup>241</sup> Am	432.2	α	3.43	α	5.37	0.115

(a) Data from ICRP 38 (1983).

(b) Includes atomic recoil and low-energy X-ray production.

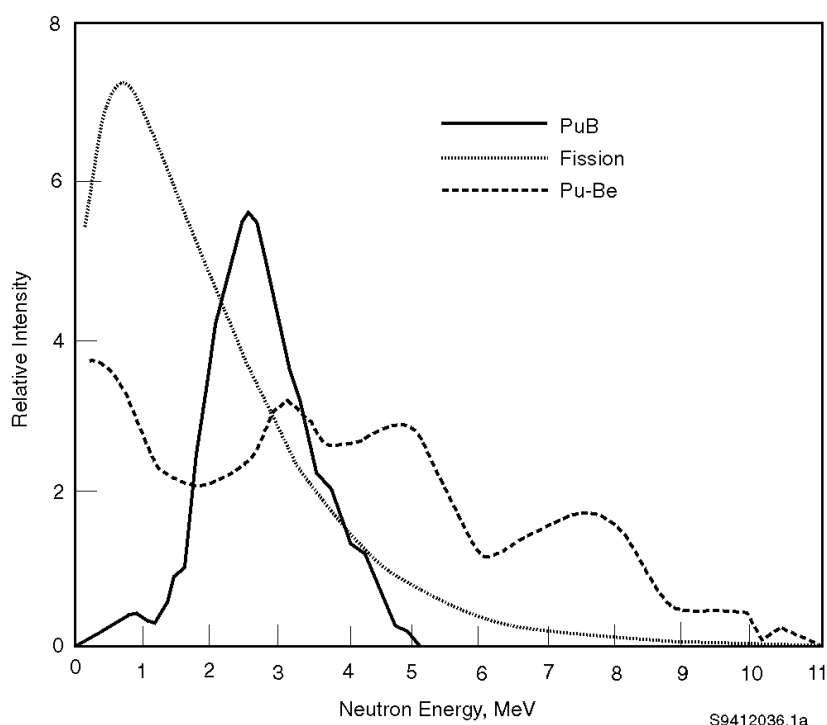
Energy spectra from Pu-Be and Pu-B neutron sources are shown in Figure 2.3. Because of licensing restrictions on plutonium, these sources have been replaced with sources fabricated from americium. Metallic plutonium emits neutrons having a Maxwellian energy distribution, with an average energy of about 1.9 MeV. Plutonium compounds and alloys also emit neutrons from alpha-neutron reactions, and these neutrons have significantly different energies:

- PuF<sub>4</sub>, about 1.3 MeV
- 10% plutonium-aluminum alloys, 1.6 MeV
- PuO<sub>2</sub>, slightly more than 2 MeV
- PuBe<sub>13</sub>, 4.3 MeV.



**Figure 2.2.** Atom Ratio of  $^{241}\text{Am}$  to  $^{241}\text{Pu}$  ( $t=0$ ) Produced by the Beta Decay of  $^{241}\text{Pu}$  as a Function of Time Since Chemical Separation

Plutonium compounds or alloys containing sodium, magnesium, silicon, chlorine, carbon, or oxygen have significant alpha-neutron yields, but little information is available about their neutron energy spectra.



**Figure 2.3** Neutron Energy Spectra of Plutonium-Beryllium and Plutonium-Boron Neutron Sources Compared with a Fission Source

## 2.3 PHYSICAL AND CHEMICAL PROPERTIES

This discussion of plutonium's physical and chemical properties begins with plutonium metal, followed by its alloys and compounds. Knowledge of the physical properties of these classes of materials and how the plutonium was produced is the key to understanding and predicting the hazards of working with this challenging element. According to Healy (1993), "Nature does not decide what happens to any material based on its radioactivity but rather on its form and mass." Form and mass are determined by the engineering application and the kinds of processes needed to achieve both intermediate and final products. Thus, to prevent nature from taking its course, there can be no shortcuts in good practices for plutonium facilities.

### 2.3.1 Plutonium Metal

The metallic state of plutonium is undoubtedly the most complicated of all the elements. Plutonium is a silvery-white metal, much like nickel in appearance. It has a low melting point (640°C) and an unusually high boiling point (3327°C). The metal exists in six allotropic forms, as indicated in Table 2.5. Two of the allotropic forms,  $\delta$  and  $\delta'$ , contract upon heating; the other forms expand upon heating. At room temperature, pure plutonium exists in the  $\alpha$  phase, which has a triclinic structure with a theoretical density of about 19.86 g/cm<sup>3</sup>. The dimensional stability of this phase is aggravated by its high linear thermal expansion coefficient and its low  $\alpha \rightarrow \beta$  transition temperature. This transformation takes place at approximately 115°C, resulting in a 10% volume change. The combination of a high specific activity and low thermal conductivity can result in significant dimensional distortion during metal-forming operations. For this reason, a  $\delta$ -stabilized dilute gallium alloy, which has a density of about 15.75 g/cm<sup>3</sup>, is used when a more dimensionally stable plutonium is desired (Merz, 1971).

**Table 2.5.** Allotropic Forms of Plutonium Metal<sup>(a)</sup>

Phase	Stability Range, °C	Density, g/cm <sup>3(b)</sup>
$\alpha$	Stable below 115	19.86
$\beta$	~115 to 200	17.70
$\gamma$	~200 to 310	17.14
$\delta$	310 to 452	15.92
$\delta'$	452 to 480	16
$\epsilon$	480 to 640	16.51

(a) Wick, 1967, p. 34.

(b) Theoretical X-ray density. The actual density is slightly lower due to crystal lattice imperfection.

Plutonium is an active metal. In moist air or moist argon, the metal oxidizes rapidly, producing a mixture of oxides and hydrides (Haschke, 1992). If the metal is exposed long enough, an olive-green powdery surface coating of PuO<sub>2</sub> is formed. With this coating, the metal is pyrophoric, so plutonium metal is usually handled in an inert, dry atmosphere of nitrogen or argon. Oxygen retards the effects of moisture and acts as a passivating agent (Raynor and Sackman, 1963). For a description of the storage hazards that the oxidation of plutonium metal creates, see Section 2.6.3.1, "Oxidation of Plutonium." A comprehensive

treatment of the oxidation of plutonium, the properties of its oxides, oxide growth, and oxidation kinetics was reviewed by Colmenares (1975).

Plutonium metal also reacts with most common gases at elevated temperatures. Plutonium metal is rapidly dissolved by HCl, HBr, 72% HClO<sub>4</sub>, 85% H<sub>3</sub>PO<sub>4</sub>, concentrated CCl<sub>3</sub>COOH (trichloroacetic acid), sulfamic acid, and boiling concentrated HNO<sub>3</sub> in the presence of 0.005M HF. The metal reacts slowly with water, dilute sulfuric acid, and dilute acetic acid. There is no reaction with the metal in pure HNO<sub>3</sub> at any concentration, with concentrated acetic acid, nor with dilute sodium hydroxide.

### **2.3.2 Plutonium Alloys**

Alloying plutonium gives rise to a host of materials with a wide range of physical, chemical, and nuclear properties.<sup>1</sup> The search for and development of new alloys has been focused mainly on the manufacture of atomic weapons, reactor fuels, heat sources, and neutron sources. The challenge of alloy development is how to maximize the desired properties without adding undesired ones. Unfortunately, some properties mutually exclude others (e.g., a gain in hardness usually results in a loss of ductility), so users may be forced to rethink their needs.

The radiological hazards of a plutonium alloy taken through its product life cycle differ from those of the pure metal isotope by virtue of the alloy's properties, which affect its form (i.e., its chemical composition, density, and geometric shape). Because form can be radically changed by external conditions (e.g., heat, pressure, and chemical atmosphere), a knowledge of the following properties will aid in evaluating the radioactive hazard:

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<sup>1</sup> See Volume 1 (Section 2) and Volume 2 (Section 5) of the *Plutonium Handbook: A Guide to the Technology* (Wick, 1967); Plutonium (Taube, 1964); and Chapter 11 of the "Reactor Handbook" in *Materials*, vol. 1 (Tipton, 1960). Beginning in 1957, a series of international conferences were held whose proceedings contain a wealth of information on plutonium alloys. From 1960 through 1975, the conferences were held every five years and produced a proceedings for each conference: Refer to *The Metal Plutonium* (Coffinberry and Miner, 1961); *Plutonium 1960* (Grison et al., 1961); "Plutonium 1965" (Kay and Waldron, 1966); "Plutonium 1970 and Other Actinides," Parts I and II (Miner, 1971); and "Plutonium 1975 and Other Actinides" (Blank and Lindner, 1976).



--	melting point	--	strength
--	diffusivity	--	ductility
--	viscosity	--	corrosion resistance
--	vapor pressure	--	pyrophoricity.

In nuclear fuel applications, the neutron absorption cross-section of the alloying elements and impurities must also be considered for its effect on radiation exposure.

### **2.3.3 Plutonium Compounds**

Much of what was said in Section 2.3.2 about the properties of plutonium alloys also applies to plutonium compounds because both are mixtures of plutonium and other elements.

Plutonium is the fifth element in the actinide series, which consists of elements with properties that stem from partial vacancies in the 5th electron shell. These elements form the seventh row in the periodic table. In general, there are four oxidation states: III, IV, V, and VI. In aqueous solutions, plutonium (III) is oxidized into plutonium (IV), which is the most stable state. The compounds  $\text{PuF}_4$ ,  $\text{Pu}(\text{IO}_3)_4$ ,  $\text{Pu}(\text{OH})_4$ , and  $\text{Pu}(\text{C}_2\text{O}_4)_2 \cdot 6\text{H}_2\text{O}$  (plutonium oxalate) are insoluble in water. The chlorides, nitrates, perchlorates, and sulfates are soluble in water. Plutonium (IV) ions complex readily with organic and inorganic compounds.

Of particular importance for radiological safety considerations are the solubility, particle size, and surface area of plutonium compounds. These properties play an important part in the transportability of plutonium in the environment and in the body. All plutonium compounds, except the oxides, are assumed in ICRP 30, Part 1 (ICRP, 1979) to behave as class W compounds in the ICRP lung model. Plutonium oxides are assumed to be class Y. The solubility of plutonium compounds is an important parameter in avoiding “unintentional” homogeneous reactors. Knowledge of this property for both aqueous and organic solvents plays a key role in criticality safety and deserves a high priority.

Unfortunately, little data on particle-size are available, and those that have been generated focus on the reactivity of the materials in the separation and conversion processes. Much of the data are reported as crystallite size, which relates to surface area and solubility but not necessarily to the way the particles would be dispersed in the air. Surface area plays a

role in the ability of materials to adsorb gases and vapors that can affect the long-term storage behavior of plutonium compounds. Pressure buildup in storage containers, either from out gassing due to self-heating or radiolytic effects, will depend on the stability of the compound and the amounts of chemisorbed or physisorbed water or other substances.

The following sections discuss the essential compounds of plutonium: plutonium nitrate and associated compounds, plutonium dioxide, plutonium hydride, plutonium sulfate, plutonium chlorides, and plutonium fuel mixtures.

### 2.3.3.1 Plutonium Nitrate, Oxalate, Peroxide, and Fluorides

Plutonium (IV) nitrate is the most used of all plutonium compounds. Essentially all chemical processing of plutonium has been conducted in nitrate solutions. These solutions of appropriate acidities range from concentrations of 10g to 250g of Pu/L for efficient precipitation processes. Intermediate compounds are also used in the processing of plutonium prepared from the nitrate: plutonium (III) fluoride, plutonium (II or IV) oxalate, and plutonium peroxide. Plutonium (IV) fluoride can be prepared from any of the preceding solids by hydrofluorination. Plutonium fluoride has been the compound of choice for reduction to the metal with calcium, principally because it is nonhygroscopic. The solubilities in various media, bulk densities, and particle sizes of these compounds are given in Table 2.6.

**Table 2.6.** Solubilities and Properties of Selected Compounds

Compound	Measured Solubility		Bulk Density, g Pu/L		Sintered Media Porosity, $\mu\text{m}^{(a)}$
	Medium	g Pu/L	Filter Cake	Dry Compound	
Fluoride (III)	1M HF - 1M HCl	0.03	-	1-2.5	15-20
Fluoride (IV)	2M HF - 2M HNO <sub>3</sub>	0.70	0.6-0.8	0.5-2.0	15-20
Oxalate (III)	0.5M C <sub>2</sub> O <sub>4</sub> <sup>2-</sup> - 3M HNO <sub>3</sub>	0.01	0.6-0.8	-	15-20
Oxalate (IV)	0.1M X <sub>2</sub> O <sub>4</sub> <sup>2-</sup> - 4M HNO <sub>3</sub>	0.003	0.5-0.6	0.6	15-20
Peroxide (IV)	3M H <sub>2</sub> O <sub>2</sub> - 1M HNO <sub>3</sub>	0.10	0.10-0.6	-	30-80

(a) Sintered media porosity required to remain precipitate.

Plutonium hexafluoride is the only volatile plutonium compound (bp 62°C) and is marginally stable. It can be prepared by oxidizing PuF<sub>4</sub> with F<sub>2</sub> at an elevated

temperature (Weinstock and Malm, 1956). It can also be prepared at low temperatures by a fluorinating agent, fluorine dioxide (Malm et al., 1984).

Plutonium waste treatment and decontamination may benefit from processes using photolysis or microwave discharge to produce active fluorine species from FOOF or  $\text{CF}_4/\text{O}_2$  mixtures, which will react with plutonium or plutonium dioxide to form  $\text{PuF}_6$  (Martz et al., 1991).

### **2.3.3.2 Plutonium Dioxide**

Plutonium dioxide may now be the most important and most thoroughly studied of all plutonium compounds. Due to its chemical stability and relative inertness, it is the preferred form for shipping and storing plutonium at the present time. Direct oxide reduction (DOR) of  $\text{PuO}_2$  is part of the integrated pyrochemical system used at the Los Alamos National Laboratory (LANL) (Christensen and Mullins, 1983; Mullins et al., 1982). Plutonium dioxide is formed when plutonium or its compounds (except the phosphates) are ignited in air, and often results when oxygen-containing compounds are heated in vacuo or in an inert atmosphere to  $1000^\circ\text{C}$  (Cleveland, 1970). The properties of  $\text{PuO}_2$  are reported by Moseley and Wing (1965).

Loose  $\text{PuO}_2$  powder, as formed by calcination, usually has a density of about  $2\text{ g/cm}^3$ . If the oxide is pressed and sintered into pellets, it may have a density of about  $10.3$  to  $11.0\text{ g/cm}^3$ . Surface measurements of typical oxides prepared from the calcination of plutonium (IV) oxalate at various temperatures range from  $10$  to  $60\text{ m}^2/\text{g}$ . Caldwell (1961) found that the surface area decreased with increasing temperatures. Plutonium oxide fired at temperatures  $>600^\circ\text{C}$  is difficult to rapidly or completely dissolve in common acids or molten salts. The best solvents are  $12\text{--}16\text{M HNO}_3$  with  $0.10\text{--}0.1\text{M HF}$ ,  $5\text{--}6\text{M HI}$ , and  $9\text{M HBr}$  (Cleveland, 1964; Holley et al., 1958). New processes are being developed to correct this deficiency using a superacid,  $\text{HF/SbF}_5$  (Olaha et al., 1985) and CEPOD, a fluoride-free electrochemical dissolver that uses the silver anion as a redox catalyst (Bray et al., 1987).

### 2.3.3.3 Plutonium Hydride

Plutonium hydride has recently become a compound of interest for separating plutonium scrap from other materials that do not readily unite with hydrogen.<sup>2</sup> The reaction between plutonium and hydrogen apparently proceeds by the initial formation of  $\text{PuH}_2$ . As more hydrogen is added, the dihydride becomes  $\text{PuH}_{2+x}$ . The hexagonal  $\text{PuH}_3$  begins to form when the H/Pu ratio becomes about 2.75; when the H/Pu ratio reaches 2.9 to 3.0, only the hexagonal form remains. A wide spread is reported in the measured induction period for the first reaction (Haschke, 1991). Because the hydriding reaction is fully reversible, plutonium metal can be recovered by pumping off the hydrogen in a suitable vacuum furnace. This metal typically contains significant amounts of plutonium oxide but is suitable for feed to either molten salt extraction or electrorefining processes. The hydride can also be converted to the oxide. The advantage of the hydride recovery process is its ability to recover a large fraction of the scrap in metallic form. This method, therefore, has a major economic advantage over chemical recycling and subsequent reduction to metal. It is being used as a production aid for metallic scrap recovery.

### 2.3.3.4 Plutonium Sulfates

Plutonium sulfate tetrahydrate,  $\text{Pu}(\text{SO}_4)_2 \cdot 4\text{H}_2\text{O}$ , has not been of any process importance but has been of interest as a primary standard for plutonium. It is a good example of a stable compound that could be suitable as an interim storage form. Samples stored at relative humidities of up to 75% showed no evidence of alpha radiolysis of the water of crystallization after 28 months. The compound is hygroscopic in air of 95% relative humidity, and stable up to 650°C, at which point it quickly decomposes to  $\text{PuO}_2$  (Cleveland, 1970). The potassium salt,  $\text{K}_4\text{Pu}(\text{SO}_4)_4 \sim 1\text{H}_2\text{O}$ , was under study as a possible primary standard for  $^{238}\text{Pu}$ . Crystals stored in an air-tight steel container, which also functioned as a heat

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<sup>2</sup> The properties of plutonium hydrides may be found in Volume 3 of the Handbook of Physics and Chemistry of the Actinides (Ward, 1985). Kinetics of the plutonium hydrogen reaction are reviewed by Haschke (1991).

sink, proved to be stable. The solubility product of this compound was determined to be  $10^{-18}$ .

#### **2.3.3.5 Plutonium Chlorides**

Chloride salts, which are a very important category of residues, are byproducts of pyrochemical operations. Pyrochemical chloride-based operations currently in use include:

- DOR
- electrorefining (ER)
- molten salt extraction (MSE)
- pyroredox.

Treatment of chloride-based residues is especially challenging for aqueous recovery techniques because of corrosion problems with stainless steel equipment. At the LANL site, Kynar-lined gloveboxes were to be installed to evaluate their behavior in production-scale operations. The Rocky Flats Plant (RFP) has also had extensive experience in aqueous recovery of plutonium from chloride-based residues (Muscatello et al., 1986a, 1986b, 1987). Cesium chloroplutonate,  $\text{Cs}_2\text{PuCl}_6$ , was proposed as a primary analytical standard due to its stability to alpha radiolysis and may now have application as a storage form. It was first prepared by Anderson (1949). There is no evidence of water absorption at relative humidities as high as 53% (Miner et al., 1963). After 64 days at 90% relative humidity,  $\text{Cs}_2\text{PuCl}_6$  forms a paste.

#### **2.3.3.6 Plutonium Fuels**

Plutonium and plutonium-uranium fuel mixtures were developed and tested in experimental reactors to prove the feasibility of operating power reactors. These fuels included both liquids and solids consisting of alloys and ceramic mixtures. Wick (1967) and Schneider and Roepenack (1986) provide comprehensive lists of fuels. Because of their pyrophoric nature, some of these alloys and

compounds require special care and handling when exposed to reactive liquids or gases.

## **2.4 RADIOLOGICAL EFFECTS ON HUMANS**

The radiobiological properties of plutonium and other transuranic (TRU) elements are known primarily from experiments performed on rats, dogs, baboons, and rabbits. Human data on plutonium are limited. Reviews of the vast literature on plutonium include Hodge et al. (1973); ICRP 19 (1972); ICRP 30, Part 1 (1979); ICRP 48 (1986); ICRP 30, Part 4 (1988b); and Liverman et al. (1974). ICRP 30, Part 1 (1979) and ICRP 48 (1986) report different gastrointestinal (GI) absorption and biodistribution parameters. The committed effective dose equivalents, calculated using the models of the two publications, will differ by about 10%, with the ICRP 48 model yielding the lesser value. Factors affecting radiobiological effects include the mode of entry of plutonium into the body, its distribution in the body, and its transfer to a fetus.

### **2.4.1 Modes of Entry into the Body**

Radioactive material can enter the body by four different pathways: by inhalation, through a wound, by ingestion, or by absorption through intact skin. These pathways may occur singly or in any combination.

- Inhalation is probably the most prevalent mode for occupational intake of plutonium. It also provides a generally conservative assumption of intake for designing bioassay programs.
- Wounds are potentially the most serious mode of intake because of the high dose-per-unit uptake of plutonium. Wounds can result from direct penetration by an object (i.e., a puncture or cut), from abrasion, or from burning by an acid, caustic, or thermal source.
- Occupational ingestion of plutonium poses a relatively small risk because the uptake factor from the GI tract to the blood is quite small and because most of the alpha energy from transformations within the GI tract is absorbed by the contents of the GI tract, rather than by the target tissues of the tract itself.

- Absorption of plutonium through intact skin is, for practical purposes, almost nonexistent. However, when removing skin contamination, care must be taken to ensure that the skin integrity is not damaged by rough or extensive decontamination procedures. If the skin integrity is damaged, the result can be considered a wound, regardless of how it occurred.

## **2.4.2 Distribution Within the Body**

Three commonly encountered biokinetic models have been promulgated by the ICRP for the internal distribution and retention of plutonium. These models are identified by the ICRP publications in which they were first reported: ICRP 30, Part 1 (1979), ICRP 48 (1986), and ICRP 30, Part 4 (1988b). The models are similar with regard to the organs of significance, but differ with regard to the fraction of uptake deposited in the organ and its respective retention (or clearance) half-time in the organ. The three models represent the ones most widely used in dosimetry and in commercially available computer codes. In all three ICRP models, once plutonium has reached the bloodstream, it is translocated primarily to the liver and skeleton. In the skeleton, it is deposited primarily on the endosteal surfaces of mineral bone, from which it is gradually redistributed throughout the bone volume by resorption and burial. Because of the extremely slow nature of this redistribution, plutonium is considered to be uniformly distributed over bone surfaces at all times following skeleton deposition. A small fraction of the translocated plutonium reaches the gonads. Although the gonadal fraction is different for males and females, the calculated gonadal doses are the same regardless of gender because the plutonium concentration in the tissues is assumed to be the same. The ICRP assumes that the remainder goes directly to excretion. Although the ICRP did not specifically state the fraction of systemic excretion occurring by urine as opposed to feces, a 0.5 fraction for each is often assumed.

Metabolic distribution and retention parameters for the three ICRP models are shown in Table 2.7. The table also includes the absorption factors from the GI tract to the bloodstream, as well as the inhalation class of common forms of plutonium.

Americium, as an ingrown impurity from the decay of  $^{241}\text{Pu}$ , can behave the same way as the plutonium host matrix in which it is contained. This implies that the  $^{241}\text{Am}$  associated with a class Y inhalation of plutonium might exhibit class Y behavior, rather than the

class W behavior assigned by the ICRP. This observation has been made in ICRP 48 (1986) and by Eidson (1980).

Experience has shown that the biokinetic models in Table 2.7 are subject to some significant variations. A Hanford plutonium-oxide-exposure case described by Carbaugh et al. (1991) has demonstrated lung retention far greater than that expected for a class Y material, leading to the suggestion of a tenaciously retained “super class Y” form. This phenomenon has been informally verified by dosimetry personnel at the Rocky Flats, Savannah River, and Los Alamos sites, and is supported in the literature by Foster (1991). At the other extreme, La Bone et al. (1992) have identified a circumstance in which a  $^{238}\text{Pu}$  oxide inhalation class appeared to exhibit biokinetic behavior more characteristic of an inhalation class D material. These extremes emphasize the importance of addressing the uniqueness of individual workers and exposure circumstances when dealing with known intakes, rather than relying on the assumed standard models.



**Table 2.7.** Common Biokinetic Models for Plutonium and Americium

Model Parameter	ICRP 30, Part 1		ICRP 48		ICRP 30, Part 4	
<b>Metabolic Distribution<sup>(a)</sup></b>	<b>F</b>	<b>T</b>	<b>F</b>	<b>T</b>	<b>F</b>	<b>T</b>
Bone surfaces	0.45	100 y	0.50	50 y	0.45	50 y
Liver	0.45	40 y	0.30	20 y	0.45	20 y
Gonads <sup>(b)</sup>						
Male	$3.5 \times 10^{-4} \alpha$		$3.5 \times 10^{-4} \alpha$		$3.5 \times 10^{-4} \alpha$	
Female	$1.1 \times 10^{-4} \alpha$		$1.1 \times 10^{-4} \alpha$		$1.1 \times 10^{-4} \alpha$	
<b>GI Tract Absorption Factor</b>						
Ox oxides	$10^{-5}$		$10^{-5}$		$10^{-5}$	
Pu nitrates	n.a. <sup>(c)</sup>		$10^{-4}$		$10^{-4}$	
Pu-others	$10^{-4}$		$10^{-3}$		$10^{-3}$	
Am-(any)	$5 \times 10^{-4}$		$10^{-3}$		$10^{-3}$	
<b>Inhalation Class</b>						
Pu oxides	Y		Y		Y	
Pu-others	W		W		W	
Am-(any)	W		W		W	

- (a) F is the fraction of plutonium reaching the bloodstream that is translocated to the organ of concern.  
T is the retention (or clearance) half-time in the organ of concern.
- (b) Plutonium is assumed to be uniformly concentrated in male and female gonadal tissue where it is permanently retained. The deposition fractions are derived, based on the relative mass of the reference male and female tissues.
- (c) n.a. = not specifically addressed.

### 2.4.3 Transfer to the Fetus

In its most recent review of the metabolism of plutonium and related actinides, it was noted in ICRP 48 (1986) that there is no strong evidence for preferential deposition of plutonium in the fetus and that the concentration of plutonium in the bone of the embryo or fetus is rapidly diluted by growth. However, experimental animal studies have shown that plutonium crosses the placenta after injection in pregnant animals (Green et al., 1979). For fallout plutonium, it has been qualitatively confirmed in humans that plutonium crosses the placenta (Okabayashi and Watanabe, 1973). However, placental and fetal membranes appear to effectively trap a portion of the plutonium that might otherwise reach the fetus.

The behavior of plutonium in the embryo/fetus changes with the development of the embryo/fetus (Sikov, 1987; Sikov et al., 1992). Liver and bone surfaces are the principal sites of plutonium deposition in the embryo/fetus, accounting for approximately 80% of the deposited plutonium (ICRP 48, 1986). Plutonium that deposits on bone surfaces

following prenatal or neonatal exposure gradually moves into the bone matrix during subsequent bone-remodeling processes.

The radiation doses produced in the embryonic stage are assumed to be relatively homogeneous and represent a small fraction of the doses received by the pregnant woman when averaged over all tissues. The dose to the fetus would constitute an even smaller fraction of the maternal dose to any tissue in which there was specific deposition (Sikov et al., 1992). As gestation progresses, there is an increase in the relative plutonium concentration in specific fetal tissues, namely the bone and liver (Sikov et al., 1992). Although limited information is available, experimental animal and human data suggest that the average concentration is higher in the fetus during the second or third trimesters than in soft tissues of the pregnant woman, exclusive of the liver, yet significantly less than in maternal tissues of primary deposition, i.e., the bone and liver.

Because placental structures, including the yolk sac, effectively trap plutonium, progenitor cells of the gametes and hematopoietic lines that appear initially in the blood islands of the yolk sac are irradiated while they are primitive stem cells. However, the dose received by the early embryonic cells and the detriment produced is not currently known.

## **2.5 RADIATION EFFECTS ON MATERIALS**

The following sections discuss, in order, self-heating and the various effects of radiolysis. Radioactive decay, particularly alpha decay, can and does affect operations in plutonium purification processes. The change in emphasis from plutonium production to waste cleanup, environmental restoration, and the retirement of nuclear weapons will present favorable circumstances for cumulative radiolytic effects, especially in the stabilization processes and the final storage form.

**Table 2.8.** Potential Hazards or Damage to Materials from Exposure to Radiation

Radiation-Induced Reaction	Potential Hazard or Damage Problem
Radiolysis of oxygen-contaminated glovebox atmospheres	Production of ozone-damage to elastomers: gloves, seals, etc.
Gaseous PuF <sub>6</sub>	Deposition of solid PuF <sub>4</sub> on equipment
PuO <sub>2</sub> exposed to hydrocarbons or humid environments	Production of hydrogen gas-pressure buildup in nonvented containers.
Ion exchange resins	Damaged resin can react violently with HNO <sub>3</sub> or other oxidizers. Also may result in hydrogen gas-pressure buildup.
CCl <sub>4</sub> saturated with H <sub>2</sub> O	Production of Cl <sub>2</sub> , C <sub>2</sub> Cl <sub>6</sub> , HCl, and phosgene.
Polyethylene	Disintegrates with production of H <sub>2</sub> .
Polyvinylchloride (PVC) plastics	Disintegrates with production of HCl-corrosion.
Tri-n-butylphosphate	Production of hydrogen and oxygen-pressure buildup in nonvented containers.
Aqueous plutonium solutions	Production of polymeric plutonium hydroxide (plutonium polymer), which plates out on vessel surfaces and piping, producing swelling, cracking, loss of ductility.
Low-acidity plutonium solutions	Increase in leachability.

Self-heating and helium retention and release are also included in this section since they too are part of the end result of the alpha decay process. Neutron production from the alpha-neutron reaction is discussed in Section 6.0. The degree of all these effects depends on the plutonium isotopic composition and the americium impurity level. Table 2.8 lists potential hazards or damage to materials from exposure to radiation.

### 2.5.1 Self-Heating

Heat generated by radioactive decay in plutonium, its alloys, or its compounds can be calculated from data provided in Table 2.4, together with the isotopic composition and plutonium fraction. The power output of reactor-produced <sup>239</sup>Pu metal is usually in the range of 2 to 10 W/kg. According to Van Tuyl,<sup>3</sup> the equilibrium surface temperature of a metal can that contains 1.2 kg of plutonium at the higher specific power would be 150°C.

<sup>3</sup> Van Tuyl, H. H. 1981. "Packaging of Plutonium for Storage or Shipment." Unpublished report by the Pacific Northwest Laboratory task force chairperson to the U.S. Department of Energy.

This calculation is complex because it depends on the thermal conductivities and configuration of all the materials in the shipping container. Thermal diffusivity measurements reported by Kruger and Robbins (1975) were combined with existing heat-capacity values to derive a curve for the thermal conductivity of the Pu-1wt% Ga alloy from room temperature to 600°C. Gram quantities of  $^{238}\text{Pu}$  can melt from self-heating under poor heat-transfer conditions. The major effects to be expected from self-heating are phase transformation, dimensional changes, chemical reactions (depending on the gaseous environment or other materials in contact with the plutonium), and desorption of previously sorbed gases or vapors.

### **2.5.2 Radiolysis**

In gases, liquids, and covalently bonded solids, the chemical effects of alpha particles and the associated recoil nucleus can cause ionization, excitation, and dissociation of molecules. From the energy requirement for ion pair formation, only about half the energy causes ionization; the other half goes into molecular excitation. Radiation effects are commonly measured by a quantity called the G-value, i.e., the number of molecules destroyed for each 100 eV of energy absorbed. For free radical production, this quantity is expressed as the  $G_R$ -value. For organic liquids,  $G_R$ -values typically range from 0.85 for carbon disulfide to 70 for carbon tetrachloride (Prevost-Bérnas et al., 1952). Although there is a considerable body of data on the radiolysis of aqueous solutions, organic liquids, and solids irradiated by gamma rays, X-rays, and fast electrons, little has been published on the radiolysis of plutonium compounds, solvents containing plutonium, or radiation-induced damage in materials that come in contact with plutonium. Nevertheless, radiation-induced damage can affect all aspects of plutonium-handling.

It would be futile and inappropriate to list, let alone discuss, all the possible radiolytic reactions affecting plutonium-handling. However, it is important to recognize the potential for and anticipate the consequences of these reactions. The following sections cover a broad range of the types of radiation-induced damage common to plutonium-handling.

### 2.5.2.1 Hydrogen Production

The G-value for the production of H<sub>2</sub> by the alpha radiolysis of pure water is 1.9±0.1 molecules of hydrogen per 100 eV (Prevost-Bérnas et al., 1952). Cleveland (1970) calculates that the energy released in 0.001M (0.24 g/L) of plutonium solution is on the order of  $2 \times 10^{14}$  eV per minute. Thus, the hydrogen evolution would be approximately  $3.8 \times 10^{15}$  molecules per liter per day for a 1M solution, or about 73 cm<sup>3</sup> of hydrogen per year.

The G-values for H<sub>2</sub> in solids irradiated by gamma rays are lower: 0.1 for ice (Johnson, 1970) and 0.01 for the hydrates of a large number of sulfates (Huang and Johnson, 1964). Because the stability of PuSO<sub>4</sub> · 4H<sub>2</sub>O was found to be remarkably high (Cleveland, 1970), one of the sulfates may well serve as an alternate interim waste form. Dole (1974) summarized the radiation chemistry of polyethylene, quoting G-values for hydrogen as 5 molecules per 100 eV. Destruction of plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). Cleveland (1970) calculated the mean change in average oxidation number in 0.5–2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI) ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species.

Pressurization of storage containers holding TRU wastes is a potential hazard for both long and interim storage periods (Kazanjian et al., 1985). Sampling of TRU waste drums shows that hydrogen is usually created (Roggenthem et al., 1989). Waste drums with pinholes can “breathe” when the atmospheric pressure changes, thereby introducing water vapor. Water vapor adsorbed on plutonium compounds is radiolytically decomposed, thereby producing hydrogen. It may be possible to add pressure relief valves and appropriate in-line filters to waste drums. (See Section 2.7 for more information on storage and containment.)

### **2.5.2.2 Redox Reactions**

In most chemical processes for purifying plutonium, it is essential to maintain its valence state. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI) ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species. Cleveland (1970) calculated the mean change in average oxidation number in 0.5-2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV.

In the radiolysis of solutions, the presence of other ionic species can accelerate or inhibit the disproportionation of plutonium valence states. For example, the presence of the chloride ion in plutonium (VI) solutions prevents reduction to plutonium (IV). Reactions may reverse after long irradiation periods, in which case a steady-state condition should ultimately be reached, resulting in a net decomposition rate of zero. An excellent review of the radiation chemistry of plutonium nitrate solutions may be found in Miner and Seed (1966). In dilute solution (0.1M),  $G_{H_2}$  is about 0.5 and  $G_{O_2}$  increases to 1.45. Self-reduction of plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). See Cleveland (1970) Chapter 2, for more information.

### **2.5.2.3 Miscellaneous Radiolytic Reactions**

A serious limitation to the use of organic ion exchange materials is their radiation stability. Brookhaven National Laboratory reviewed the literature and summarized the effect of ionizing radiation on both organic and inorganic ion exchange materials (Gangwer et al., 1977). Extraction of plutonium (IV) from 3M  $HNO_3$  into 30 vol% tributyl phosphate in kerosene at 5°C decreased the extraction coefficient by a factor of two when irradiated to a dose of  $3.6 \times 10^7$  R (Tsujino and Ishihara, 1966). The mechanical properties of thin plastic films such as polyethylene and polyvinyl chloride degrade with exposure to plutonium. Cellulose vacuum-cleaner bags will disintegrate in less than a month if used for housekeeping purposes in plutonium-contaminated gloveboxes.

Leachability of plutonium-containing wastes could be affected by the production of nitric acid for air-equilibrated dilute salt solutions (Rai et al., 1980).

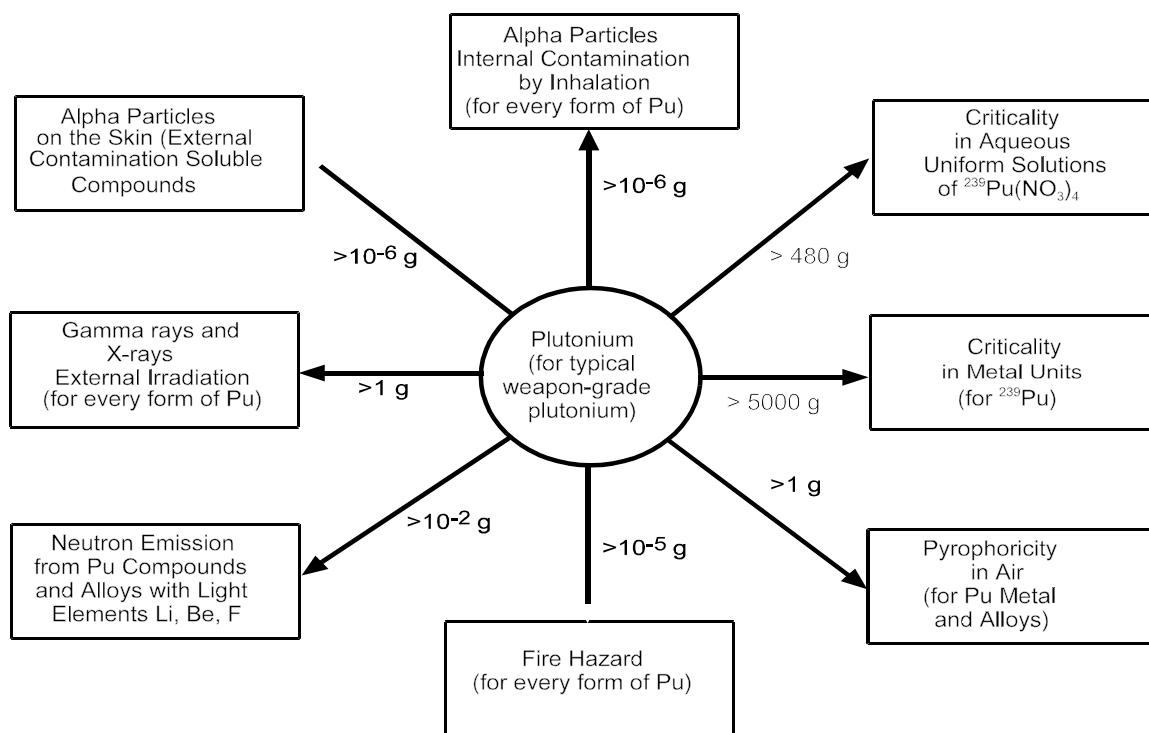
#### **2.5.2.4 Helium Retention and Release**

Helium introduced by alpha-bombardment of plutonium and the alloys and compounds of plutonium can cause lattice expansion. This was first observed for plutonium oxides, carbides, and nitrides by Rand et al. (1962) and was later observed for two plutonium carbide phases. Helium is retained in vitrified compounds. The retention and release behavior of helium in plasma-torch-fused  $\text{PuO}_2$  microspheres for SNAP is an important parameter in the design of the heat source. Approximately  $530 \text{ cm}^3$  at standard temperature and pressure (STP) per year-kg are produced by  $^{238}\text{PuO}_2$  (Stark, 1970). Microspheres of 80%  $^{238}\text{PuO}_2$  and 20%  $^{239}\text{PuO}_2$  that were approximately 50 mm in diameter, prepared by the sol-gel process, released 92.8% of the helium in 8 months at room temperature (Northrup et al., 1970). Metals at temperatures well below the melting point trap the insoluble helium gas in tiny bubbles, which are more or less evenly distributed through the matrix material (Stevens et al., 1988). Helium buildup in weapon-grade material is approximately 4 standard  $\text{cm}^3$  per year-kg.

## **2.6 OCCUPATIONAL HAZARDS**

The major industrial hazard in plutonium facilities is the potential for loss of control of a highly toxic substance, resulting in either the inhalation or ingestion of plutonium or one of its compounds by personnel, or the exposure to excessive radiation from a criticality accident. The possibility of a fire or explosion in a plutonium facility is probably the most serious threat because the consequences of a fire could lead to loss of containment and subsequent dispersement of highly mobile plutonium particulates. In addition, fighting the fire with water to maintain containment could create the potential for a criticality accident and/or loss of containment in the immediate vicinity.

The day-to-day hazards for personnel in plutonium facilities involve exposure to gamma rays, X-rays, and neutrons, as well as possible accumulation of plutonium in the body. These hazards are described in more detail in Section 3.0, "Radiation Protection," and Section 7.0, "Nuclear Criticality Safety." The amount of plutonium needed to present potential hazards to personnel in plutonium-handling facilities is summarized in Figure 2.4. Hazards related to interim and long-term storage of plutonium will be found in Section 2.7, "Storage and Containment."



**Figure 2.4.** Hazards in Low-Exposure Plutonium Handling

### 2.6.1 Chemical Versus Radiological Hazards

The radiological toxicity of reactor-produced plutonium far exceeds the chemical toxicity of this heavy element. Furthermore, its low solubility in near-neutral or basic solutions reduces the uptake through ingestion by a factor  $>1000$  for any plutonium compounds except certain complexes, such as the citrate or ethylenediamine tetraacetic acid (EDTA)



complex. (Refer to Sections 2.3, “Radiobiological Properties,” and 6.0, “External Dose Control”). Tipton (1960) summarizes the differences in chemical hazards between plutonium and uranium: “In contrast to uranium, the chemical toxicity of plutonium is insignificant in comparison to the hazard arising from its natural radioactivity.” Moreover, “the toxicity of plutonium and other transuranic elements,” according to Voelz et al. (1985), “has only been studied in animals since acute toxicity has never been observed in man for these elements and epidemiologic studies have not produced positive results.” However, recent evidence suggests that plutonium can catalyze reactions including oxidative stress in the absence of significant radioactive decay. These data presented by Claycamp and Luo (1994) suggest that plutonium complexes might contribute to long-term oxidative stress related to tumor promotion.

## **2.6.2 Associated Chemical Hazards**

The main chemical hazard of plutonium is its vulnerability to oxidation and the pyrophoricity of some of its alloys and compounds (see Section 2.6.3).

The processing of plutonium, including separation from irradiated uranium, purification, conversion, waste disposal, environmental restoration, and D&D, necessarily requires the use of chemicals and reagents with varying degrees of toxicity and hazardous properties. A partial list of chemicals that are either used or proposed for use at DOE plutonium facilities is provided in Table 2.9. An abbreviated evaluation of the potential hazards of these substances is also provided. Table 2.9 is not meant to replace the Material Safety Data Sheet (MSDS) available from chemical manufacturers; rather, it is intended to help readers recognize the toxicity of these chemicals and identify any possible side effects from their use that could jeopardize radiation safety or plutonium containment.

## **2.6.3 Hazards Created by Oxidation and Pyrophoricity**

This section describes the oxidation and burning characteristics of plutonium, summarizes the storage properties of the metal and oxides, and presents recommendations for their storage conditions. Waste remediation plans for TRU materials and the necessity for dealing with ton quantities of plutonium metal from the retirement of weapons require the identifying of long-term and intermediate-term waste forms with

appropriate stability. Economic considerations make clear the importance of generating few, if any, new wastes in accomplishing this task.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities***Table 2.9.** Hazards of Chemicals Used in Processing Plutonium<sup>(a)</sup>

Chemical Name	CAS No. <sup>(b)</sup>	Hazard	Formula
Aluminum nitrate	7784-27-2	—	AL(NO <sub>3</sub> ) <sub>3</sub> 9H <sub>2</sub> O
Antimony pentafluoride	7783-70-2	—	SbF <sub>5</sub>
Beryllium (metal)	7740-41-7	Neutron	Be
Calcium (metal)	7740-70-2	Releases H <sub>2</sub> when wet, flammable	Ca
Calcium oxide	1305-78-8	Corrosive	CaO
Calcium chloride	10043-52-4	—	CaCl <sub>2</sub>
Ferrous ammonium sulfate	7783-85-9	—	Fe(SO <sub>4</sub> )(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> 6H <sub>2</sub> O
Fluorine	7782-41-4	Oxidizer, poison	F <sub>2</sub>
Fluorine dioxide	—	Oxidizer, poison	F <sub>2</sub> O <sub>2</sub>
Carbon tetrachloride	56-23-5	—	CCl <sub>4</sub>
Ferrous sulfamate	—	—	Fe (SO <sub>3</sub> NH <sub>2</sub> ) <sub>2</sub>
Gallium (metal)	7440-55-3	—	Ga
Hydrogen	1333-74-0	Flammable, explosive	H <sub>2</sub>
Hydrogen fluoride	7664-39-3	Corrosive	HF
Hydrochloric acid	7647-01-0	Corrosive	HCl
Hydrogen peroxide	7722-84-1	Oxidizer	H <sub>2</sub> O <sub>2</sub>
Iodine	7553-56-2	Poison	I <sub>2</sub>
Magnesium (metal)	7439-95-4	Water reactive, flammable, explosive, produces neutrons when combined with Pu	Mg
Magnesium chloride	7786-30-3	Neutron	MgCl <sub>2</sub>
Magnesium oxide	1309-48-4	Neutron	MgO
Mercuric nitrate	10045-94-0	Oxidizer, poison	Hg(NO <sub>3</sub> ) <sub>2</sub>
Nitric acid	7697-37-2	Oxidizer, corrosive, poison	HNO <sub>3</sub>
Oxalic acid	144-62-7	Poison	H <sub>2</sub> C <sub>2</sub> O <sub>4</sub>
Potassium hydroxide	1310-58-3	Corrosive, poison	KOH
Potassium chloride	7447-40-7	—	KCl
Sodium chloride	7647-14-5	—	NaCl
Sodium hydroxide	1310-73-2	Corrosive, poison	NaOH
Sodium nitrate	7631-99-4	Oxidizer	NaNO <sub>3</sub>
Sodium nitrite	7632-00-0	—	NaNO <sub>2</sub>

**Table 2.9.** (continued)

Chemical Name	CAS No. <sup>(b)</sup>	Hazard	Formula
Stannous chloride	7772-99-8	—	SnCl <sub>2</sub>
Sulfamic acid	5329-14-6	Corrosive	NH <sub>2</sub> SO <sub>3</sub> H
Sulfuric acid	7664-93-9	Corrosive, poison	H <sub>2</sub> SO <sub>4</sub>
Tri-n-butyl phosphate	126-73-8	Flammable liquid	(C <sub>4</sub> H <sub>9</sub> O) <sub>3</sub> PO <sub>4</sub>
Urea	57-13-6	—	CO(NH <sub>2</sub> ) <sub>2</sub>
Uranium (metal)	—	Flammable	U
Zinc chloride	7646-85-7	—	ZnCl <sub>2</sub>
Soltrol 170 Phillips 66	68551-19-9	Flammable liquid isoparaffins)	(Mixture C10–C14
Carbon tetrafluoride	75-73-0	—	CF <sub>4</sub>

(a) Refer to Material Safety Data Sheets for complete discussion of hazards.

(b) Chemical Abstracts Service Registry number.

### 2.6.3.1 Oxidation of Plutonium

The problems of oxidation of metallic plutonium were recognized shortly after the discovery of plutonium, and extensive studies of the low-temperature corrosion of plutonium and its alloys have been performed. Oxidation can produce fine loose plutonium oxide, which disperses easily in glovebox systems, complicating housekeeping chores. If not controlled, loss of accountability and increased radiation exposure to personnel is certain. The reactivity of plutonium metal is discussed in Section 2.3.1. The tendency for enhanced oxidation is promoted by the self-heating properties of plutonium isotopes (discussed in Section 2.5.1). A kilogram of <sup>239</sup>Pu can easily reach an equilibrium temperature of 80°C in a glove-box environment (Raynor and Sackman, 1967). Thermally isolated <sup>238</sup>Pu metal can easily melt from its own decay heat. The heat generated by oxidation may be sufficient to ignite nearby combustible materials. Metal turnings and scrap should be reprocessed or converted to stable alternatives as soon as practicable. Plutonium metal, its alloys, and its reactive compounds need to be excluded from both oxygen and water vapor, but especially the latter since it catalyzes and accelerates oxidation.

The corrosion or oxidation of plutonium does not always occur in a linear or predictable manner. The oxidation rate is a complex function of the surrounding atmosphere, the moisture content, and the alloys or impurities present in the metallic plutonium.<sup>4</sup>

### **2.6.3.2 Ignition Temperatures and Pyrophoricity of Plutonium, Its Alloys, and Its Compounds**

Plutonium and some of its alloys and compounds are pyrophoric. Pyrophoric material is a liquid or solid that, even in small quantities and without an external ignition source, can ignite within 5 minutes after coming in contact with air (NFPA Fire Protection Handbook). Pyrophoric plutonium metal has been defined as “that metal which will ignite spontaneously in air at a temperature of 150°C (320°F) or below in the absence of external heat, shock, or friction” (Stakebake, 1992).<sup>5</sup> Finely divided plutonium metal would be considered pyrophoric while massive plutonium would be nonpyrophoric. Martz et al. (1994) has proposed a mechanism for plutonium pyrophoricity that predicts the ignition temperature as a function of surface mass ratio and particle size.

The most numerous forms of pyrophoric plutonium are chips, lathe turnings, and casting crucible skulls. Plutonium hydride and sesquioxide ( $\text{Pu}_2\text{O}_3$ ) are probably the most commonly occurring pyrophoric compounds. Plutonium carbide, oxycarbide, nitride, and oxide phases with compositions between the sesquioxide and dioxide are potentially pyrophoric. Known pyrophoric alloys include Pu-U and Pu-Ce. Waber (1967) summarized much of the early work on plutonium corrosion and oxidation and is a good source for identifying other pyrophoric alloys.

The health physics aspects of an accidental plutonium fire can be serious. A fire can burn through containment structures, resulting in the dispersal of  $\text{PuO}_2$  over a

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<sup>4</sup> See Wick (1967), Coffinberry and Miner (1961), and Kay and Waldron (1966) for details on the oxidation of unalloyed plutonium and the stabilized alloy of plutonium.

<sup>5</sup> Also in DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

wide area, with the potential for inhalation exposure during the fire or during subsequent decontamination efforts. The conditions under which a plutonium fire can occur in a dry glovebox have been studied. With only 5% oxygen in nitrogen, the metal will burn easily. At the 1% level, however, a fire will not continue to burn unless heat is supplied (Rhude, 1962). Turnings must be generated in a dry atmosphere and should be converted to the oxide as soon as convenient, preferably on the same day they are made. Some solvents and organic compounds form flammable mixtures with plutonium. In one incident, tetrachloroethane was inadvertently substituted for another lathe coolant in a metal-turning operation. Chips of plutonium aluminum alloys were ignited, resulting in the blowout of a glove-box panel. In a separate event, burning plutonium chips dropped into carbon tetrachloride resulted in an explosion (AEC, 1965).

#### **2.6.3.3 Aerolization of Plutonium**

The ignition of plutonium metal becomes a major hazard when enough plutonium has burned to produce a significant amount of dispersable material and a serious enough fire to damage the pertinent containment structures. The particle size of  $\text{PuO}_{1.9}$  fired at a low temperature varies from 3% at  $<1\ \mu\text{m}$  to 97% at  $1\text{--}5\ \mu\text{m}$  (Stakebake and Dringman, 1967). Sintered  $\text{PuO}_{2.0}$  has a particle size  $<2\ \mu\text{m}$ . Haschke (1992) made an effort to define the maximum value of the source term for plutonium aerosolization during a fuel fire. He found the rate to be constant ( $0.2\text{-g PuO}_2/\text{cm}^2$  of metal surface per minute) above  $500^\circ\text{C}$ . The mass distribution for products of all metal gas distributions are approximately 0.07 mass% of the oxide particles having geometric diameters  $\leq 10\ \mu\text{m}$ .

## **2.7 STORAGE AND CONTAINMENT**

The DOE mission for utilization and storage of nuclear materials has recently changed as a result of the end of the "Cold War" era. Past and current plutonium storage practices largely reflect a temporary, in-process, or in-use storage condition which must now be changed to accommodate longer-term storage.

The DOE has sponsored a number of workshops on disposing of plutonium. Two of the objectives of these workshops have been to make recommendations for near-term and long-term storage forms and to identify possible alternatives. At the Hanford Plutonium Disposition Workshop held in Richland, Washington, from June 16 to 18, 1992, the two highest ranking stabilization processes were, first, denitration of plutonium nitrate, and, second, thermal stabilization. The third-ranked process included the precipitation of  $\text{Cs}_2\text{PuCl}_6$  or  $\text{K}_4\text{Pu}(\text{SO}_4)_4$  followed by thermal stabilization (Hoyt, 1993). At the workshop on plutonium storage sponsored by DOE Albuquerque, on May 26 and 27, 1993, both metal and oxide were considered suitable storage forms. A report has been issued summarizing information presented here and resulting from this workshop (DOE, 1994a). This important report includes sections on:

- materials properties relevant to storage;
- current storage practice (DOE Facilities, RFP, LANL, Hanford, SRS, and ANL);
- advanced storage concepts;
- hazard analysis; and
- recommendations.

A report entitled "Technical Issues in Interim Plutonium Storage" by J. C. Martz, J. M. Haschke, and M. C. Bryuson, LANL, submitted to Arms Control and Nonproliferation Technologies, attempts to provide a technical basis for addressing complex interfaces with political and economic issues. Its goal is to identify alternative storage options for excess plutonium. Currently, DOE is circulating Draft Interim Recommendations for Storage of Plutonium Metal and Plutonium Oxide at Department of Energy Facilities. The principal difference between interim and long-term storage is the need for transfer of plutonium from a contaminated glovebox environment into an improved, hermetically sealed storage container without the inclusion of plastic or other organic materials. Existing storage and handling requirements for plutonium metal and oxides are currently covered in DOE Order 0460.1A and DOE Order 6430.1A (DOE, 1996b and 1989b).

The following property summaries adapted from Haschke and Martz (1993), are useful for determining potentially unsuitable storage and containment conditions for plutonium metal and oxide. Given that plutonium metal is chemically reactive in air and other environments, it also:

- Exhibits spontaneous self-sustained ignition (becomes pyrophoric) only if the metal dimension
  - is  $<0.1$  mm and  $T > 150^{\circ}\text{C}$
  - is  $>0.2$  mm and  $T > 500^{\circ}\text{C}$
- reacts slowly in air at room temperature (maximum of about  $1\text{ }\mu\text{m/day}$ )
- has limiting (T-independent) oxidation rate in air above  $500^{\circ}\text{C}$
- is not a dispersible form ( $<10\text{ }\mu\text{m}$  geometric size) until oxidation occurs:
  - oxide from Pu+Air at ambient T: 100 mass % (ssa =  $10\text{--}20\text{ m}^2/\text{g}$ )
  - oxide from  $\text{PuH}_2 + \text{O}_2$ :  $\sim 25$  mass % (ssa  $\sim 1\text{ m}^2/\text{g}$ )
  - oxide from  $\text{Pu} + \text{O}_2$  and Pu+Air at  $T > 500^{\circ}\text{C}$ :  $< 0.1$  mass % (ssa  $< 0.1\text{ m}^2/\text{g}$ )
- radiolytically decomposes organic and covalently bound specific species in the environment
- reacts with most radiolytically produced gases and with nonequilibrium surface:
  - limits pressurization by gases
  - forms low-density (pressure-generating) and pyrophoric products
- retains helium from alpha decay
- is stabilized by certain storage atmospheres (reactivity decreased by  $10^{12}$ )
- is stable if isolated from reactive species
- has good storage history when stored properly.

A similar property summary for plutonium dioxide, the most commonly used form of plutonium, shows it to be stable and unreactive in air. Storage and containment recommendations, based on the properties of plutonium metal and dioxide, are shown in Table 2.10.



**Table 2.10.** Storage Recommendations for Plutonium Metal and Dioxide (adapted from Haschke and Martz, 1993)

- 
- Metal and oxide are both suitable storage forms for plutonium (100 years).
  - Organics (plastics, elastomers) must be excluded from the primary container for both forms.
  - Converting between metal and oxide is not recommended (negative impact of waste, cost, environmental safety and health [ES&H] risk).
  - Both forms must be properly prepared and certified:
    - Procedures for metal already exist (technology transfer needed).
    - Procedures for oxide need development (stabilization, desorption, loss on ignition [LOI]).
  - Both forms must be in sealed primary containers for extended storage:
    - Positive seals (e.g., welds and metal seals) are necessary.
    - Seal certification or double sealing is necessary.
  - Requirements diverge for short-term/retrievable storage:
    - Containers with metal gaskets are advantageous for metal storage.
    - After stabilization, oxide is best stored in a container fitted with a rupture disk in series with a vented stainless-steel frit container.
  - Surveillance of stored materials is required.
-

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### 3.0 RADIATION PROTECTION

The radiation protection field is concerned with the protection of individuals, their progeny, and humanity as a whole, while still allowing for necessary activities which might involve radiation exposure. The aim of radiation protection is to prevent deterministic effects and to limit the probability of deterministic effects. Most decisions about human activities are based on an implicit form of balancing risks and benefits leading to the conclusion of whether or not the application of a particular practice produces a positive net benefit. Because the probability of health effects is not zero, the ICRP in Publication 26 (ICRP, 1977) recommended the following criteria for a system of dose limitation:

- No practice shall be adopted unless its introduction produces a positive net benefit.
- All exposures shall be kept as low as reasonably achievable, with economic and social factors being taken into account.
- The dose equivalent to individuals shall not exceed the limits recommended for the appropriate circumstances.

These criteria and related information have been incorporated into DOE regulations, instructions, and manuals for radiation protection.

The successful operation of a plutonium facility requires scrupulous attention to providing adequate radiation protection and maintaining contamination control through the implementation of a quality health physics program. (In this section, “health physics” and “radiation protection” can be used interchangeably when referring to programs or personnel.) Prompt dose assessment is important for demonstrating compliance with standards, providing information to workers, establishing an accurate historical record, and for responding to accident and incident situations. This section defines the basis for the establishment of a sound health physics program at a plutonium facility.

### **3.1 REGULATION AND STANDARDS**

Regulations on radiation protection in DOE and DOE contractor facilities are found in 10 CFR 835, Occupational Radiation Protection: Final Rule (DOE, 1993c). Guidance is found in the supporting document Radiological Control Manual (DOE, 1994l) and Implementation Guides.

Certain OSHA regulations, primarily those contained in 29 CFR 1910 (DOL, 1993), may also be included in the radiation protection program for workers in DOE facilities. Other related source documents include publications of the EPA, ANSI, ICRP, NCRP, and UNSCEAR.

In addition, each site that handles radioactive materials and/or radiation generating machines, is required to establish and maintain its own documented radiation protection program, following the Federal regulations.

### **3.2 RADIATION PROTECTION PROGRAMS**

Radiation protection programs include provisions for quality assurance, administrative controls, protection of visitors, visits by regulatory personnel, and onsite packaging and transportation of hazardous materials.

#### **3.2.1 Quality Assurance**

It is highly desirable for laboratories and industrial facilities handling plutonium to have a well-integrated quality assurance program. Such a program should have high visibility and strong management support. Quality assurance should be effectively applied throughout facility activities, including the radiation protection program. The basis for quality assurance programs in DOE facilities is established in 10 CFR 830, Nuclear Safety Management (DOE 1994p) and DOE Order 5700.6C, Quality Assurance (DOE, 1991a). In addition, 10 CFR 830.120, Quality Assurance Requirement, requires the development of a Quality Assurance Program, specifies an implementation schedule, and provides the elements that the program must address.

An effective quality assurance program for radiation protection will include establishment of appropriate standards of performance for essential activities and equipment, with an effective system of documentation and traceability of those activities

and of the use of the equipment. Proper maintenance of those records will be necessary for reference purposes.

### **3.2.2 Administrative Controls**

In any facility that handles radioactive materials, the major controls protecting workers, the public, and the environment are structures and installed equipment, which shield, contain, and confine the radioactive materials. However, to allow useful work to be performed in the facility and to assure that its protective features remain effective, a number of administrative controls are ordinarily required. These administrative controls are usually contained in a series of procedures related to the operations and maintenance activities to be carried out in the facility. All personnel who work in controlled areas should be familiar with the administrative controls that apply to their work. When changes or additions to administrative controls are made, these changes or additions should be effectively communicated to all persons who may be affected.

#### **3.2.2.1 Radiation Protection Procedures**

A plutonium facility should have a written policy on radiation protection, including a policy on keeping exposures ALARA. All radiation protection procedures and controls should have formal, recognizable technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to account for all copies and ensure that all new procedures are included in the historical files. A designated period of time for maintaining historical files should be established. DOE Order 200.1 (DOE, 1996a) and ANSI N13.6-1989 (ANSI, 1966) provide guidance on how to maintain historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur. Guidance in writing procedures can be found in DOE/NE/SP-0001T, Writer's Guide for Technical Procedures (DOE, 1991b).

Radiation protection procedures should be provided for but not limited to the following topics:

- Posting and labeling of facilities
- development and maintenance of all radiation protection records
- reporting of unusual radiation occurrences
- use of radiation monitoring instruments
- use of radiation sources (e.g., reference calibration)
- reporting of radiation exposures
- use of protective clothing
- responding to radiological emergency events
- surveying and monitoring
- counting room equipment and use
- instrument maintenance and control
- development and use of Radiological Work Permits (RWPs)
- responsibilities of operations staff for contamination control and personnel surveys.

Two topics, RWPs and facility posting and labeling, are discussed below in more detail.

### **3.2.2.2 Radiological Work Permits**

Radiological Work Permits or other work planning documents should be used for entry into high and very high radiation areas, high contamination areas, and airborne radioactivity areas. The RWPs should also be used to control entry into radiation and contamination areas and for handling materials with removable contamination. The RWPs should be initiated by the work group responsible for the activity. All RWPs should be reviewed and approved by the radiation protection staff. Radiological Work Permits are recommended for other radiological work in accordance with the Radiological Control Manual (DOE, 1994I). Most plutonium facilities will use them for all but routine entries into rooms containing glove-box lines. Guidance for posting of RWPs and for their contents are contained in the Radiological Control Manual.

Radiological workers should read and understand the applicable RWP before performing work in a radiological area. The RWPs should be located at the access point to the applicable radiological work area. Workers should acknowledge by signature or through electronic means that they have read,

understood, and will comply with the RWP before they initially enter the area and after changes. Out-of-date RWPs should be removed.

### **3.2.2.3 Radiological Surveys and Data Trending**

Area monitoring in the workplace shall be routinely performed, as necessary, to identify and control potential sources of personnel exposure (10 CFR 835.401(b)). This monitoring should include surveys in areas that are not ordinarily expected to be contaminated. The program should define minimum requirements, survey types, and frequencies.

Surveys should be performed at frequencies adequate to identify changes in posting required or an activity buildup, and to ensure that current radiological controls are appropriate. The surveys suggested by this section are minimum recommendations; additional surveys should be conducted, recorded, and reviewed as necessary to ensure full protection of personnel.

Contamination surveys should be performed to determine surface contamination area (SCA) boundaries, the appropriate posting of sources or areas, and the location and extent of localized contamination.

Contamination surveys should be performed and documented prior to the start of radiological work, during general work activities at times when changes in contamination level may occur, and following work to assure that final radiological conditions are acceptable and documented. See Munson et al. (1988).

A sufficient number of points should be surveyed to adequately assess the radiological status of the area being surveyed.

Routine radiological surveys should be regularly conducted, recorded, and reviewed for all areas where personnel could be exposed to alpha, beta, gamma, X-ray, or neutron radiation throughout the site. Surveys should be performed at frequencies adequate to ensure protection of personnel. The following surveys should be considered the minimum. Additional surveys should be conducted,

recorded, and reviewed as necessary to ensure that personnel exposures are maintained ALARA.

General radiation and contamination surveys should be performed:

- To identify and verify the boundaries of areas which must be radiologically controlled
- to verify that radiation and contamination levels in uncontrolled areas remain less than specified limits
- to determine the appropriate posting of localized higher radiation levels, beams, or hot spots
- to ensure that radiological conditions are acceptable and documented prior to, during, and at the completion of work that may cause changes in radiation levels to occur (see Munson et al., 1988, p. 6.1.2)
- to satisfy required predetermined procedure hold-points in work areas and adjacent areas, whenever operations are performed that may cause significant increases in radiation levels. The survey may be required as part of a radiological inspection step required by the work procedure. This includes areas above and below the work area as appropriate during special processing operations or cell decontamination, movement of permanent or temporary shielding, radioactive waste processing, and relocation of highly radioactive materials.

Routine radiation and contamination level surveys should be performed in the workplace at a frequency commensurate with the radiation hazard, to detect trends related to equipment, systems, environment, and work habits.

Nonroutine surveys of radiation and contamination levels in the workplace should be performed:

- Before initial use of a new installation, system, or equipment, or as soon as possible after a radiation source is brought into the area



- whenever changes in procedures, equipment, or sources have occurred that may cause changes in the external radiation levels
- after modification to a shield or changes in shield materials
- as the basis for trend evaluation of external radiation level conditions
- when a radiological accident has occurred or is suspected
- when requested by the personnel performing the activity (see Munson et al., 1988, p. 6.1.2).

Radiation surveys should be performed upon initial entry into process cells and tanks that contain radioactive piping or components.

Surveys should be conducted when performing operations that might result in personnel being exposed to small intense beams of radiation (e.g., removing shielding for shielded X-ray devices).

Every reasonable effort should be made to maintain the radiation dose equivalent of the surveyor at levels that conform to ALARA guidance.

Surveys should be performed and documented according to established procedures.

Only fully trained and qualified personnel should conduct surveys that are to be the official records of radiation levels or for the protection of personnel; these surveys should be reviewed and approved by the Radiological Protection Manager or his/her designee.

Surveys should be performed with calibrated instrumentation appropriate for the intensity and energy of the radiation anticipated in the area to be surveyed.

Survey instruments should meet the performance check requirements established by the facility in accordance with ANSI N323 (ANSI, 1993).

Combinations of survey instruments should be used as necessary to provide the capability to measure all types of radiation and dose rates characteristic of that which could be encountered at the facility being surveyed.

Records that establish the conditions under which individuals were exposed to external radiation (such as facility radiological conditions records generated by the monitoring programs) should be retained to provide a chronological and historical record. See ANSI-N13.6-1989 (ANSI, 1966).

A sufficient number of points should be surveyed in order to adequately assess the radiological status of the area. Regular predetermined points may be used, but additional spot monitoring should be done to ensure that all changes in dose rates are identified, recorded, and reviewed.

All records of surveys should clearly identify, as a minimum:

- The name, signature, and employee number of the surveyor
- survey instrument(s) model number, serial number, and calibration date
- the type(s) of radiation being monitored (e.g., neutron, gamma, etc.)
- the dose rates
- the date and time the survey was performed
- locations where radioactive material is located temporarily (or is being temporarily stored) or where equipment that generates ionizing radiation is being operated.

Records of the results of radiation surveys should be retained in accordance with facility policy.

Survey data should be reviewed by supervisory personnel. Significant findings should be presented to the facility manager in a timely manner.

Health physics personnel should summarize survey data in each building or area at least once a quarter. Significant changes or trends in area dose rates and/or radiological contamination should be noted and corrective actions assigned. The survey summary should be presented to the facility management quarterly.

Survey results and data summaries should be made available to the ALARA team chair periodically and should be used:

- To provide a basis for evaluating potential worker exposure on a job and in ALARA preplanning
- to provide a baseline for trend analysis, investigation, and correction of unusual conditions
- to track the status of jobs (including identification of good practices) and to detect departures from good operating procedures and/or the failure of radiation controls
- to identify the origin of radiation exposures in the plant by location, system, or component.

Health physics personnel should post the results of radiation surveys or survey maps at the entrance to all permanent radiation areas, high radiation areas, and very high radiation areas. The results should be posted in the form of a survey map so that personnel can be aware of the locations of higher and lower levels of radiation within the area.

A survey data trending program should be conducted; to indicate the continuing effectiveness of existing control; to warn of deterioration of control equipment or effectiveness of operating procedures; to show long-term variations in radiation levels; and to identify and correct improper radiation work practices. See NUREG-0761 (NRC, 1981) 07.B(I)(C), 09.B(4), and 09.C(I)(C).

Health physics should perform trend analyses on all permanent radiation, high radiation, and very high radiation areas. At a minimum, one complete survey record should be evaluated and included in the trend analysis program for each

survey required to be performed by the facility routine control program. See NUREG-0761 (NRC, 1981), 07.B(I)(C), 09.B(4), and 09.C(I)(C).

Health physics should use the facility reporting system to identify discrepancies and abnormal trends and should summarize the data review results in their monthly reports to the Radiological Protection Manager.

Survey data trends should be investigated when either:

- an upward trend in general area radiation level occurs, causing a significant increase
- an abrupt change in radiation level occurs that cannot be directly correlated to normal activities.

#### **3.2.2.4 Facility Posting and Labeling**

Areas in plutonium facilities shall be posted in accordance with the requirements in 10 CFR 835 (DOE, 1993c). Implementation Guide G-10 CFR 835/G1-Rev. 1, Implementation Guide. Posting and Labeling for Radiological Control (DOE, 1994e) provides guidance to ensure compliance. The technical criteria and dose rate and/or levels for defining radiation, high radiation, very high radiation, contamination, high contamination, and airborne radioactivity areas are established in 10 CFR 835. The health physics staff should identify:

- Areas to be barricaded and marked to prevent personnel from inadvertently entering them
- Areas to be physically controlled per 10 CFR 835, Subpart F

Entrance to radiological areas shall be controlled (10 CFR 835.501(a and b)) commensurate with the existing and potential radiological hazard within the area.

The health physics staff should post current radiation surveys of radiation areas at the health physics access control point for use in prejob planning. Airborne Radioactivity Areas shall be posted with the words, "Caution, Airborne

Radioactivity Area” when the airborne radioactivity levels in the occupied area exceed, or are likely to exceed, 10 percent of the DAC value listed in Appendix A or Appendix C of 10 CFR 835 (10 CFR 835.603(d)). These areas are posted to alert personnel of possible respiratory protection requirements.

#### **3.2.2.5 Unposted Areas**

Certain areas of facilities that handle radioactive materials should be maintained free of detectable radioactive contamination. These areas should also be maintained at ambient radiation levels equivalent to the environmental background of the facility. Parts of the facility that should meet these requirements include lunchrooms, offices, restrooms, janitor rooms, corridors outside operational areas, foyers, and outside areas surrounding the facility, including the building roofs.

To assure these areas meet the requirements of radiological cleanliness, they should be surveyed with count-rate instruments sensitive to the radioactive isotopes of interest. In a plutonium facility, the instruments should meet the requirements listed in ANSI Standard N317-1991, Performance Criteria for Instrumentation Used for In-Plant Plutonium Monitoring (ANSI, 1980a). These clean areas should be maintained below the surface contamination levels cited in 10 CFR 835 (DOE, 1993c).

#### **3.2.3 Visitors**

Regardless of the general radiation safety knowledge of visitors to a plutonium facility, they should be escorted at all times when they go into the posted areas of the plant. In addition, before going into such an area, they should be given a general orientation to the facility radiation protection program and informed about the potential radiation conditions in the areas where they will be going. They also should be provided with the same protective devices worn by facility personnel engaged in similar activities.

Visitors with a demonstrated need to enter the following areas may be allowed access if such access is controlled with a combination of training and the use of escorts trained for the specific area:

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- Radiological Buffer Areas
- Radiation and High Radiation Areas
- Contamination Areas
- Radioactive Material Areas.

Guidance for training for visitors is provided in the Radiological Control Manual (DOE, 1994l), Articles 622 and 657:

- Persons under 18 years of age should not be permitted to enter Radiation Areas or Contamination Areas without the approval of the Radiological Protection Manager.
- Area entry requirements and access restrictions for visitors should be in accordance with established facility procedures.
- Individuals, visitors included, shall (10 CFR 835.502(b)) be prevented from entering Very High Radiation Areas when dose rates are in excess of the posting requirements of 10 CFR 835.603(c), and visitors should be prohibited from accessing High Contamination and Airborne Radioactivity Areas.

In addition the following is recommended:

All facility personnel serving as a qualified escort should ensure that each visitor under his/her cognizance completes a facility radiological visitor form. The qualified escort should also sign the visitor form and complete it as appropriate.

Facility-sponsored visitors should provide the following before entering radiological areas, unless these records have already been entered into the facility entry control system:

- Evidence of completing required training, as applicable
- visitor radiation exposure disclosure

The host facility manager should forward the visitor radiation exposure and medical disclosure forms to Dosimetry.

The use of offsite mask fit certification may be authorized (if in accordance with the applicable Radiation Protection Program) under the following conditions:

- A mask fit has been completed within the previous year.
- The individual presenting the mask fit certification card has not changed physical appearance in a way that would affect the seal of the mask to the face.
- The facility has the masks available that the individual is certified to wear.

### **3.2.4 Visits by Regulatory Personnel**

Periodically, personnel from the DOE and other Federal and state agencies visit radiation facilities for audit purposes or to discuss regulatory changes. In most cases, they will want to look at records of the radiation protection program and, in some cases, will also want to enter posted areas of the facility. They should have ready access to the facility provided that dosimetry and other requirements are met. They should have complete access to facility personnel knowledgeable in the subjects they wish to discuss.

### **3.2.5 Onsite Packaging and Transportation**

The hazardous materials organization conducts onsite radioactive shipments with the assistance of health physics. This program requires the hazardous materials organization representatives to review onsite radioactive shipping records, document the errors or omissions observed, and evaluate trends and revise training as needed. Serious deficiencies are to be documented and the reports should be submitted in accordance with DOE Order 460.1A, Packaging and Transportation Safety (DOE, 1996b).

Onsite packaging systems for shipments of radioactive material are generated to fulfill a need of a user organization.

Generally, an assessment is required for each onsite package containing Type B, fissile, or highway route control quantities (HRCQ).

The packaging organization is responsible for coordinating onsite package design and preparation of safety analysis documentation. The following information describes typical process, review, and approval requirements for onsite safety analysis documentation:

1.       **Initiation.** New safety analysis documentation or reviews/changes to existing documentation can be requested by a user organization based on programmatic or operational requirements. The request is submitted in writing to the packaging organization and includes proper justification and support documentation. The packaging organization makes routine revisions as necessary to reflect policy and regulation changes.
2.       **Preparation.** The packaging organization coordinates the analysis, prepares safety analysis documentation, and guides the documentation through the review and approval process, including the resolution of review comments and the obtaining of required approval.
3.       **Control.** Safety analysis documentation is prepared and maintained according to facility policy. The document control system provides an accessible, auditable, and retrievable method for maintaining and changing safety analytic documentation.
4.       **Review and Approval Cycle.** Safety analysis documentation is reviewed, approved, and changed according to facility policy. Additional reviews and approvals include the following people and organizations:



- User
- cognizant engineer
- packaging organization
- quality assurance
- responsible environmental assurance organization, onsite only
- packaging, shipping, and waste safety assurance organization
- criticality engineering analysis, if criticality analysis is required
- packaging and shipping approval authority
- DOE field office, if the package is to be used for HRCQ inter-area shipments.

5. **Approval for Editorial Changes.** Inconsequential editorial changes to a safety analysis document may be approved at the operating level.

6. **Utilization.** Once a safety analysis document is approved, copies are sent to the affected organizations, including operations and applicable facility engineering, to incorporate the administrative controls from the safety analysis document into the affected operating documents. User organizations must obtain the packaging organization review of all operating procedures that incorporate instructions or administrative controls found in COCS, SARPS, SEPS, DAPS, DOT exemptions, and Federal and state packaging requirements to ensure that they are properly incorporated.

Onsite packages currently approved for onsite use should be cataloged and described in a hazardous materials packaging directory maintained by the packaging organization. New packages are added to the directory as they are developed and approved.

### 3.3 RADIOLOGICAL CONTROL ORGANIZATIONS

The radiological control organization must be structured so that all of the activities required to provide support to line management and workers can be accomplished.

### **3.3.1 Management Commitment**

Management commitment to safety is the most important characteristic of an effective radiological control program. If the management commitment to safety is strong, the radiological control program will be valued and respected. The radiological control program should be provided adequate authority to permit performance of necessary assignments and program implementation. Management commitment to the ALARA concept is particularly important [see Article 111, Radiological Control Manual (DOE, 1994)]. Adequate personnel, equipment, and funding should be available as a part of this commitment.

### **3.3.2 Radiological Control Organization Independence and Reporting Level**

The radiological control organization should be independent of the line organization responsible for production, operation, or research activities and should have an equivalent reporting level. Because health physics personnel should have the authority to balance operations with safety, they should not report directly to the administrators of operations. When shift work is involved, the operations shift supervisor may make minor health physics decisions in support of the shift's Radiological Control Technicians (RCTs); however, decisions involving basic policies and procedures should be directed to a separate health physics organization.

If a safety organization includes the health physics program, it should be high enough in the company to assure direct access to the company president or equivalent. If the health physics program is administered by a separate radiological control organization, that organization should also be in a position to assure direct access to the company president. This is to safeguard the program from the pressures of production that exist in the operational environment, by keeping it independent of operating organizations.

A system of guides, policies, and procedures should be established to clearly identify the interrelationships, responsibilities, and authorities of those involved with the development, operation, and maintenance of the facility and the health and safety of the employees. These guides, policies, and procedures should be documented and should be reviewed at least once every year.

### **3.3.3 Adequacy of Personnel and Equipment**

A sufficient number of qualified and, where required, certified radiological control personnel should be available to perform necessary tasks for support of plutonium facility startup and operation (See Section 3.4 for guidance concerning staffing and staff qualifications). Sufficient equipment, including protective clothing, respiratory protective equipment, and radiation detection instrumentation should be available to support RCTs and operating personnel in the performance of work in controlled areas.

### **3.3.4 Assignment of ALARA Responsibility and Authority**

Limiting radiation exposures to the lowest levels commensurate with the benefit of the work to be accomplished has long been a part of health physics and radiological protection programs of DOE and its contractors. 10 CFR 835 (DOE 1993c) establishes the policy of maintaining ALARA exposures of workers and the public to radiation from DOE operations. Procedures are required to be prepared (10 CFR 835.1003(a)) and implemented and records must be maintained (10 CFR 835.701) to demonstrate the implementation of ALARA. The DOE Radiological Control Manual (DOE, 1994l) provides additional guidance. Munson et al. (1988) and G-10 CFR 835/B2-Rev. 1, Implementation Guide. Occupational ALARA Program (DOE, 1994c), may be used in developing an ALARA program.

An ALARA committee should be established at the plutonium facility. The membership should include managers and workers from the line, the technical support organization, and the radiological control organization. A line manager, such as Director of Operations, Research, or Maintenance should serve as the committee chair. The ALARA committee should make recommendations to management to improve progress toward minimizing radiation exposure and radiological releases (DOE, 1994a).

## **3.4 STAFFING AND STAFF QUALIFICATIONS**

A cadre of operating and maintenance personnel that has experience in the operation of a plutonium facility should be established during the construction of a new facility. The remainder of the operating and maintenance staff should be hired as soon as possible and should receive formal and informal training from the experienced personnel. This step is extremely important to enable all personnel to grow with the facility and learn the details of the operations. Once operations start,

potential problems already should have been identified and engineering or administrative changes should have been made to resolve them.

Staffing in the radiological control organization requires technicians and professionals in many support areas. A successful health physics program is highly dependent on the availability of adequate staff support in areas such as environmental monitoring, instrument maintenance and calibration, internal and external dosimetry, meteorology, safety analysis, and risk management.

#### **3.4.1 Professional Staffing and Qualifications**

The senior staff of the radiological control organization should include health physicists and other professionals with four-year degrees in science or engineering. A continuing training program should be established for facility personnel. Pursuit of certification by the American Board of Health Physics for senior and professional staff members is encouraged (DOE, 1994a).

At least one professional staff member at the plutonium facility should have a minimum of three years of health physics experience in the operation of plutonium facilities.

#### **3.4.2 Technician Staffing and Qualifications**

Recommendations for minimum entry-level requirements for RCTs are given in the DOE Radiological Control Manual (DOE, 1994l) and DOE/EH-0262T, Radiological Control Technician Training (DOE, 1995f). They include a high school education or equivalency and knowledge of certain scientific fundamentals. If a two-year degree in nuclear technology or an equivalent discipline is locally available, completion of such a program should be encouraged.

Where possible, the Radiological Control Techniques (RCTs) and other members of the health physics staff should have a minimum of one year's experience working at a plutonium facility. Such experience is an important prerequisite to allowing them to work unsupervised. Personnel hired without such experience should work an internship of six months under the leadership of a qualified RCT or supervisor with experience in that facility.

The RCTs should be encouraged to pursue registration by the National Registry of Radiation Protection Technologists.

### **3.4.3 Staffing Levels**

At least one professional health physicist is recommended to be on the staff of each major plutonium facility as a full-time employee.

There is no rule of thumb for determining the number of RCTs needed for a given plutonium facility. The number of RCTs should be based on an analysis that provides for sufficient coverage on each shift, given the number of samples, surveys, and other work to be performed; the time of training, donning and doffing protective clothing; shift turnover procedures; and other similar considerations. The site collective dose and individual dose limits in the facility may also lead to the need for additional personnel. Consideration should be given to having sufficient personnel to respond to off-normal conditions and emergencies as well as routine work. Major maintenance, modifications, or decommissioning activities may require additional personnel.

## **3.5 INSTRUMENTATION CONSIDERATIONS**

The radiation from the radioactive decay of plutonium includes alpha, beta, gamma, X-ray (photons), and neutron radiation. An effective monitoring program for plutonium requires radiation detection instruments that are responsive to all of these forms of radiation. It is essential that instruments meet the performance criteria outlined in the applicable U.S. and international standards and be properly calibrated for their intended use.

### **3.5.1 Types of Instruments and Measurements**

Alpha-sensitive instruments are necessary for most contamination control surveys. Exposure rate surveys are normally conducted with photon-sensitive instruments with known energy responses. Neutron surveys become important when processing tens of grams of  $^{238}\text{Pu}$  or hundreds of grams of mixed isotopes of plutonium, particularly compounds (i.e.,  $\text{PuO}_2$ ,  $\text{PuF}_4$ , etc.). The neutron survey is important in instances where photon shields, such as leaded glass, are used; such shields normally stop all of the charged particles, most of the low-energy photons, and essentially none of the neutrons. Under these circumstances, neutron radiation is likely to be the major contributor to whole body dose.

Continuous air monitors (CAMs) are used extensively in plutonium facilities. Continuous air monitors and sample extraction lines that go to CAMs and continuous radiation dose monitors should be placed outside the glove boxes and hoods. In-line processing instrumentation is critical to accurately monitor the work stations and a review should be performed to determine instrument locations. Continuous air monitors may not have adequate detection capabilities for real-time monitoring at the DAC level. For  $^{239}\text{Pu}$ , the annual limit on intake (ALI) is 4.8 nCi for class W compounds based on the DAC of  $2 \times 10^{-12} \mu\text{Ci/mL}$ , as given in Appendix A to 10 CFR Part 835 (DOE, 1993c). Representative manufacturers' specifications on the performance level of such a CAM range from 1 DAC in 4 hours (4 DAC-h) to 1 DAC in 8 hours (8 DAC-h) for alarm (with no radon present). Continuous air monitors typically have had poor large-particle response due to particle loss during transport to the filter inside the system. Newer alpha air monitors are able to handle large particles more efficiently. Background levels of radon-thoron decay products may be present in concentrations up to 50 to 100 times greater than the level of plutonium of interest. If calibrated properly, alpha CAMs will subtract background levels of radon-thoron decay products; however, in practice the detection limit for plutonium may be as high as 40 DAC-h in the presence of high radon levels. A new generation of alpha CAMs, just coming onto the market, is able to compensate for radon more effectively and meet the desired 8 DAC-h alarm level.

Transuranic aerosol measurement units have been developed and adapted to be used in the workplace. These units avoid preferential plate-out of larger particles by using an in-line filter. Higher flow rates than those normally used with CAMs may be used. Increased detection is obtained on a quasi-real-time basis by high-volume air sampling and counting in a separate vacuum chamber. Detection levels of less than 0.5 DAC-h have been quoted for these units. It has been demonstrated that high-volume impact samplers used at some facilities have demonstrated detection capabilities of 0.1 DAC-h in the laboratory and 1 DAC-h in the field. Other monitoring systems that use diffusion, impaction, or electronic discrimination to reduce the effect of background resulting in an increased detection capability have also been used and are being improved upon. However, it is suggested that site-specific testing be performed on any new equipment to ensure compatibility and verify expected performance. See the Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) for additional information on the selection, placement, and operation of plutonium air monitors.

### **3.5.2 General Performance Criteria for Instruments**

Programs for in-plant monitoring of plutonium consist mainly of airborne and surface contamination surveys and dose rate surveys. The general and specific performance criteria for the instrumentation needed to conduct these programs are described in ANSI N317-1991 (ANSI, 1980a). Performance specifications are also given in ANSI N323-1993 (ANSI, 1993), ANSI N42.17A (ANSI, 1988b), and ANSI N42.17C-1989 (ANSI, 1987c) for portable health physics instrumentation and IEC Publication 325 (IEC, 1981) for alpha and beta contamination meters and monitors. Criteria for air monitoring instrumentation are contained in ANSI N13.1-1993 (ANSI, 1969b), IEC Publication 761-2 and draft IEC Publication 761-6 (IEC, 1983), and ANSI N42.17B-1989 (ANSI, 1987b). Criticality alarm systems are discussed in ANSI/ANS 8.3-1986 (ANSI, 1986a). The criteria discussed in the following subsections are specified in these standards as referenced.

### **3.5.2.1 Portable Survey Instruments**

ANSI N317 (ANSI, 1980a) discusses several criteria related to the performance of portable survey instruments; these include the following requirements:

- The overall accuracy shall be within  $\pm 20\%$ , and the precision shall be within  $\pm 10\%$  at the 95% confidence level.
  - The response time (i.e., the time for the instrument reading to go from zero to 90% of full scale) shall be  $< 10$  seconds on the most sensitive scale and  $< 2$  seconds at readings of 100 mrem/h, 100 mR/h, and 500 dpm or greater. (This criterion is unrealistic with current neutron instrument capabilities. Response time is typically 30 to 60 seconds.)
  - The instrument shall be able to maintain accuracy and precision for a minimum of 24 hours of continuous operation.
  - The instrument shall have a minimum battery lifetime of 200 hours of continuous operation.
- ANSI N42.17A (ANSI, 1988a) specifications differ slightly.
- The response of the instrument shall not change by more than  $\pm 15\%$  from a reference value taken at  $20^{\circ}\text{C}$  over the anticipated temperature range for operation.

- The instrument system shall function within specifications over all anticipated combinations of temperature and humidity (e.g., 15° to 65°C, 40% to 95% relative humidity).

ANSI N317 (ANSI, 1980a) states that the minimum detection capability for **alpha survey instruments** ideally should be 220 dpm/100 cm<sup>2</sup> of surface area and shall not be more than 500 dpm/100 cm<sup>2</sup>. This requirement is to be met in the presence of a radiation field of 0.10 rem/h of neutrons in the energy range of thermal to 10 MeV, and/or in the presence of 0.10 rem/h of photons in the energy range of 0.010 to 1.25 MeV. The operating range should be from 0 dpm to at least 100,000 dpm/100 cm<sup>2</sup> of surface area. The response of the instrument to beta-interfering radiation is an important specification that is to be stated by the manufacturer.

**Photon survey instruments** should meet the accuracy requirements stated in ANSI N317 (ANSI, 1980a) over the energy range of 0.01 to 1.25 MeV. The angular response of this type of instrument should be within  $\pm 15\%$  over a  $2\pi$  steradian frontal direction using at least two photon sources with energies ranging from 0.06 to 1.25 MeV. Experience has shown that this response specification is not met by most instruments at lower energies due to attenuation of the photon. The energy dependence should be within  $\pm 15\%$  over the range of very low energy to 1.25 MeV and the operating range should be from 0.5 mR/h to at least 5000 mR/h. Experience has shown that  $\pm 20\%$  over very low energy to 1.25 MeV is more realistic. This specification applies to a specific window selection (e.g., below 0.05 MeV, the electron equilibrium cap or beta shield must be removed).

According to ANSI N317 (ANSI, 1980a), the response of **neutron survey instruments** for neutron energies in the range of thermal to 10 MeV shall approximate the dose equivalents given in that standard for instruments that are designed for dose equivalent rate measurements. The angular response for neutron instruments should be within  $\pm 15\%$  in a  $2\pi$  steradian frontal direction for <sup>252</sup>Cf energy neutrons or equivalent. The operating range is to be from 0 to at least 2000 mrem/h.



ANSI N42.17A (ANSI, 1988b) has a broader scope than ANSI N317 (ANSI, 1980a) but the criteria in it apply to portable survey instruments. Additional criteria include geotropism (maximum change of 6% from reference reading for all orientations), temperature shock, mechanical shock, vibration, and ambient pressure (maximum change of 15% from reference reading for the latter four criteria). Some differences exist between ANSI N42.17A and ANSI N317. In most cases, the criteria for ANSI N42.17A are more applicable because these criteria are based on substantial testing, which was sponsored by DOE. In ANSI N42.17A, precision is tied into a measurement level; for example, it quotes a precision of 15% at <500 cpm and 10% at >500 cpm. Also, with the advent of liquid crystal displays and other digital readouts, "response time" is defined as the time it takes for the reading to move from 10% to 90% of the equilibrium or steady-state reading. Another significant difference in the standard is that the battery lifetime specification is 100 hours instead of the 200 hours mentioned in ANSI N317.

For direct alpha contamination surveys, the use of audible signals (headphones or speaker) greatly facilitates the detection of "hot spots."

IEC Publication 325 (IEC, 1981) provides additional guidance on the uniformity of probe response for alpha and beta contamination meters. Surface sensitivity measurements are also discussed in this standard.

**3.5.2.2 Performance Criteria for Fixed Monitoring Instruments**

Airborne contamination monitors, surface contamination monitors, photon and neutron area monitors, and emergency instrumentation are fixed monitoring instruments subject to the following standard performance criteria.

**Airborne Contamination Monitors.** Airborne contamination monitors, normally CAMS (see Section 3.5.1), should meet the following criteria according to ANSI N317 (ANSI, 1980a). The primary purpose of any CAM is to detect the presence of airborne radioactivity and activate an alarm to warn personnel in the area so that actions can be taken to minimize personnel exposures. The goal for any CAM should be to perform this function as quickly as possible and at the lowest detectable level of radioactive airborne concentration. The quantity of airborne radioactivity that will result in an alarm within a given time interval is defined in units of DAC-h for a particular radionuclide and is a function of the nuclide's airborne concentration in DACs, the sampling rate, the lower limit of detection of the instrument, and the time needed for the alarm to occur. Mishima et al. (1988) provides guidance on each of these functions.

The minimum detection level of  $^{239}\text{Pu}$ , in terms of derived air concentration (DAC), should be 8 DAC-h at the point of sampling in the presence of nominal amounts of naturally occurring alpha-emitters such as radon and thoron and their decay products. (No guidance is provided on what a "nominal" amount is, however.) The operating range should be at least 100 minimum detection levels (i.e., up to 800 DAC-h for  $^{239}\text{Pu}$ ). Instrument error should not exceed  $\pm 20\%$  of the reading over the upper 80% of the operating range. The reproducibility of the system for any given measurement should be within  $\pm 10\%$  at the 95% confidence level for a mid-scale or mid-decade reading. The instrument should be capable of operating with less than a 5% change in calibration over the ambient temperature range expected. The instrument should be equipped with an adjustable alarm set point (audible and visible alarms) that can be set at any point over the stated range. The air flow rate should be indicated and adjustable. Voltage and frequency variations of  $\pm 15\%$  within design values should result in reading variations of no greater than 5% at the minimum detection level.

ANSI N42.17B (ANSI, 1987b) provides additional performance criteria for air monitors used to detect plutonium. This standard provides specifications for general criteria (sampler design, units of readout, alarm threshold, etc.), electronic criteria (alarms, stability, response time, coefficient of variation, and line noise susceptibility), radiation response, interfering responses (radiofrequency, microwave, electrostatic, and magnetic fields), environmental criteria (temperature, humidity, and pressure), and air-circuit criteria. More detailed specifications are provided in ANSI N42.17B than in ANSI N317 (ANSI, 1980a); however, the environmental criteria and the limits of variation are not as restrictive as those in ANSI N317. With respect to accuracy, ANSI N317 requires less than  $\pm 20\%$ , and ANSI N42.17B requires 40% at the 95% confidence level. For the environmental criteria, ANSI N317 requires that the readings change less than 5% under ambient conditions, while ANSI N42.17B gives a 15% limit of variation. As discussed previously, criteria from ANSI N42.17B are more applicable because they are supported by instrument testing.

ANSI N13.1 (ANSI, 1969a) provides detailed guidance on sampling methods. One criterion that relates to CAMs is that air sample lines between air inlet and filter media are to be eliminated where possible; where not possible, they are to be designed to meet the sampling criteria contained in the standard (e.g., short lines, proper sampling rate, smooth bends). The use of Tygon tubing as sample lines before the collection filter should be minimized or eliminated. Air in-leakage from surrounding areas can be a problem when using sampling lines. Testing for air in-leakage shall be performed at least annually or when seals or "O" rings are replaced.

**Surface Contamination Monitors.** Surface contamination monitors include hand and/or shoe counters and instruments (or probes) with sufficient flexibility to survey pieces of equipment, including exterior clothing. ANSI N317 (ANSI, 1980a) states that these instruments shall have an audible alarm, a frequency that is proportional to the count rate, or a preselectable trip setting, and that upon reaching that level shall activate an audible or visible alarm or both. These instruments should be calibrated according to the requirements in ANSI N323 (ANSI, 1993) and be equipped with a traceable check source. Fixed instruments should be powered by alternating current (AC) and provided with an emergency power source.

**Photon and Neutron Area Monitors.** Photon and neutron area monitors measure the intensity of photon and neutron radiation in areas where significant quantities of plutonium are stored and/or handled. ANSI N317 (ANSI, 1980a) states that these monitors shall have a preselectable trip setting with audible annunciators, shall provide electronic signals for remote alarms if they are used as alarming devices, and shall be equipped with a visual meter or digital readout. All neutron and photon area monitors should be AC-powered and all critical monitors should be provided with an emergency power source. Many of the requirements that apply to portable survey instruments, as stated in ANSI N317 may also apply to this type of instrumentation. Calibrations should be performed according to the requirements in ANSI N323 (ANSI, 1993).

### 3.5.2.3 Performance Criteria for Emergency Instrumentation

Meeting the criteria for criticality accident alarm systems, fixed nuclear accident dosimeters, and other emergency instrumentation is essential.

**Criticality Alarm Systems.** ANSI/ANS 8.3 (ANSI, 1986a) discusses the performance and design criteria for criticality accident alarm systems. The criteria include the following:

- Criticality alarm systems shall be designed to detect immediately the minimum accident of concern; the minimum accident may be assumed to deliver the equivalent of an absorbed dose in free air of 20 rad at a distance of 2 meters from the reacting material within 60 seconds.
- Systems shall be designed so that instrument response and alarm latching shall occur as a result of radiation transients of 1-millisecond duration. The alarm signal shall be for evacuation purposes only and of sufficient volume and coverage to be heard in all areas that are to be evacuated. Very high audio background noise in some areas may require that the alarm be supplemented with visual signals; however, high background noise is a dangerous situation that should be prevented by design. Instrument response to radiation shall be calibrated periodically to confirm the continuing performance of the instrument. The calibration interval may be determined on the basis of experience but shall be no

less frequent than annually. Tests should be performed at least monthly and the results of testing should be documented.

The standard does not quantify criteria for reliability or the rejection of false alarms. Consideration should be given to the avoidance of false alarms as accomplished by providing reliable single detector channels or by requiring concurrent response of two or more detectors to initiate the alarm. (ANSI 1986a).

**Fixed Nuclear Accident Dosimeters.** All DOE facilities that have sufficient quantities and kinds of fissile material to potentially constitute a critical mass, such that the excessive exposure of personnel to radiation from a nuclear accident is possible, shall provide nuclear accident dosimetry for those personnel (10 CFR 835.1304). Requirements for fixed nuclear accident dosimeters are found in DOE Order 420.1 (DOE, 1995c).

**Effluent Monitors.** Facilities that deal with unencapsulated plutonium should have continuously operating effluent monitors to determine whether or not plutonium is being released to the environment. Effluent monitor criteria is found in IEC Publications 761-1 and 761-6 (IEC, 1983) and ANSI N42.18 (ANSI, 1974b) and should be performed. Similar to airborne contamination monitors, effluent monitors should be tested for air in-leakage at least annually or when seals or "O" rings are replaced.

**Other Emergency Instrumentation.** Other emergency instrumentation should provide ranges for all radiation dose rates and contamination levels potentially encountered at the time of an accident. Normally, dose rate capabilities from a few millirem per hour to a few hundred rem per hour should be required while capability requirements for the contamination level may range upward from 200 dpm/100 cm<sup>2</sup> for alpha contaminants and 100 dpm/100 cm<sup>2</sup> for beta-gamma emitters. Performance specifications for emergency radiological monitoring instrumentation can be found in ANSI N320-1979 (ANSI, 1975b) and BNWL-1742 (Andersen et al., 1974).

### **3.5.3 Instrument Calibrations and Testing**

Radiation doses and energies in the work areas should be well characterized. Calibration of instruments should be conducted where possible under conditions and with radiation energies similar to those encountered at the work stations. Knowledge of the work area radiation spectra and instrument energy response should permit the application of correction factors when it is not possible to calibrate with a source that has the same energy spectrum. All calibration sources should be traceable to recognized national standards. Neutron energy spectral information is considered particularly important because neutron instruments and dosimetry are highly energy-dependent.

When the work areas have been well characterized, the calibration facility used by the plutonium plant should be set up to represent as closely as possible the work area's radiation fields. Californium-252 or PuBe calibration sources should be used for work areas that process plutonium metal and plutonium oxide because their neutron energy distribution is similar to those compounds. Facilities that process PuF<sub>4</sub> should use a PuF<sub>4</sub> source. Most work areas at processing plants are high-scatter areas and thus have significant quantities of low-energy neutrons. Because it may not be feasible to have sources and scatter geometries representative of all work locations at the facility, it should be important to determine specific spectra and correction factors for work locations to correct for the calibration. Scatter conditions should be taken into account when setting up a calibration facility. The effect of room scatter in a neutron calibration facility can be significant and may account for as much as 20% of the measured dose equivalent rate. The Schwartz and Eisenhauer (1982) methods should be used to correct for room scatter.

ANSI N323 (ANSI, 1993) provides requirements on the calibration of portable instruments. The reproducibility of the instrument readings should be known prior to making calibration adjustments. This is particularly important if the instrument has failed to pass a periodic performance test (i.e., the instrument response varies by more than  $\pm 20\%$  from a set of reference readings using a check source) or if the instrument has been repaired. The effect of energy dependence, temperature, humidity, ambient pressure, and source-to-detector geometry should be known when performing the primary calibration. Primary calibration should be performed at least annually.

Standards referenced in Section 3.5.2 discuss specific performance testing of radiation detection instruments. Testing procedures in these standards should be used for periodic requalification of instruments or detailed testing of instruments.

The calibration of photon monitoring instruments over the energy range from a few keV to 300 keV is best accomplished with an x-ray machine and appropriate filters that provide known x-ray spectra from a few kiloelectron volts to approximately 300 keV. Radionuclide sources should be used for higher energies. Most ion chambers used to measure photon radiations have a relatively flat energy response above 80 to 100 keV;  $^{137}\text{Cs}$  or  $^{60}\text{Co}$  are typically used to calibrate these instruments. These sources also may be used to calibrate Geiger-Mueller (GM) type detectors used for dose rate measurements. It should be noted that some GM detectors (e.g., those with no energy compensation) can show a large energy dependence, especially below approximately 200 keV. GM detectors should not be used if not energy compensated.

The calibration of alpha-detection instruments normally should be performed with  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ , or  $^{230}\text{Th}$  sources. Several sources of different activities should be used to calibrate different ranges.

Whenever possible, beta detectors should be calibrated to the beta energies of interest in the workplace. A natural or depleted uranium slab source can be used for calibration of beta detectors when beta radiations in the workplace have energies similar to the uranium. International Organization for Standardization beta sources should be used for all other purposes: the energy dependence of beta detectors can be tested using the calibration sources listed in the ISO Publication 1980 (1984); these include  $^{90}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{204}\text{Tl}$ , and  $^{147}\text{Pm}$ .

The calibration and testing of crucial monitoring systems are extremely important to the overall radiation protection program but have often been neglected. Effluent monitoring and sampling systems and remote area monitoring systems (RAMs) should be given several tests. The radiological, environmental, and mechanical characteristics of the instrumentation portion of the system should be fully evaluated prior to its first use to ensure its compatibility with performance requirements and facility operating conditions. The effluent sampling losses from the sample probe to the collector/detector should be determined. This test should be repeated at least annually and when a significant change in the sampling equipment is made. The sample probe should be examined at least once a year to verify that its design or performance has not been changed by corrosion. The recorder of the sample

flow rate should be calibrated when it is installed and annually thereafter. The operability of the overall system should be completely tested once, with repeat tests only after modification, repair, or maintenance. Operability checks should be scheduled at least monthly and calibration performed at least annually.

The operation of criticality or other radiation alarm signal systems should be checked periodically to ensure that the alarms are audible at all potentially occupied locations. To prevent any desensitizing of staff, the staff should be aware that the tests will be performed, and where possible, tests should be scheduled during off-shift hours. Building systems should be tested semiannually and the area-wide system should be tested at least annually. Any portion of the detector/alarm system that is affected by the test should be reconfirmed for operability after the test is completed (e.g., if a detector is disconnected and a signal is injected at that point, the detector should be tested immediately after it has been reconnected).

### **3.6 RADIATION SAFETY TRAINING**

A thorough radiation protection training program should be established at plutonium facilities. Separate training programs should be established for general employees, radiation workers, and RCTs. The training of all staff members should be carefully documented. The DOE Radiological Control Manual (DOE, 1994l), and DOE standardized training programs (DOE, 1992c and DOE, 1995g) provide guidance on information to be presented during the training programs.

The frequency requirements for Radiation Safety Training are specified in 10 CFR 835.901. Refresher training in the alternate year when retraining is not performed is recommended. Individuals who work with plutonium should have special plutonium facilities training, such as DOE/EH-0425 Plutonium Facilities Training (DOE, 1994k) in addition to Radiological Worker Training (DOE, 1995g).

Training requirements must ensure that personnel have the training to work safely in and around radiological areas and to maintain their individual radiation exposure and the radiation exposures of others ALARA.

#### **3.6.1 Radiological Worker Training**



Before working in plutonium operations, all radiological workers shall be trained and qualified according to 10 CFR 835.902. A thorough radiation protection training program should be established at plutonium facilities. Before beginning plutonium training, each plutonium worker should receive Radiological Worker Training.

The level of radiation worker training should be determined in accordance with the Radiological Control Manual, Table 6.1 (DOE, 1994l). All training should be in accordance with Radiological Worker Training DOE/EH-0260T-1 (DOE, 1995g) and implemented by the guidance of G10 CFR 835/J1, Rev. 1, Implementation Guide. Radiation Safety Training (DOE, 1994f). All training dispositions and records shall be documented in accordance with 10 CFR 835.704 (DOE, 1993c).

### **3.6.2 Radiological Control Technician Training**

A thorough RCT training program should be established at plutonium facilities. Before plutonium operations begin, a trained and qualified staff of RCTs should be present. All RCT training should be accomplished in accordance with DOE EH-0262T, Radiological Control Technician Training (DOE, 1995f).

### **3.6.3 Training for Other Facility Personnel**

Nonradiological workers in a plutonium facility should be given a general orientation on the radiation safety concerns for working with plutonium, the general protective measures used for work with plutonium, and the engineered safety features of the facility.

### **3.6.4 General Public Education**

If there are members of the public who live or work near a plutonium facility, a plan for orientation of members of the public should be developed to inform them of facility activities. Such a plan should include information on the concerns that require protection of people from potential injuries by plutonium, the general protective measures used at the facility to confine it and keep it out of the public domain, and solicitation of information on the concerns of members of the local public about plutonium. To the extent possible, efforts should be made to allay those concerns. The information in the public education plan should also be provided to local news media.

### **3.6.5 Training Qualifications**

All training instructors and materials should meet the requirements in DOE Order 5480.20A (DOE, 1994j) and should meet the guidance in the Radiological Control Manual (DOE, 1994l).

Each plutonium facility should develop performance-based training that reflects radiological conditions present at the facility. This training should be monitored to ensure that site-specific, worker-performance-based measures, and practical factors are included in the plutonium training.

### **3.6.6 Health Physicist Training Involvement**

Facility health physicists should have comprehensive knowledge of all of the material on plutonium radiation safety that is included in the training programs for radiation workers and RCTs.

## **3.7 RADIOLOGICAL RECORDS**

The systematic generation and retention of records relating to the occupational radiation protection program are essential to describe the occupational radiation exposure received by workers and the conditions under which the exposures occurred. Such records have potential value for medical, epidemiological, and legal purposes.

Regulation 10 CFR 835 (DOE, 1993c) establishes radiation protection program records requirements. The Radiological Control Manual (DOE, 1994l) provides guidance for radiation protection program records.

10 CFR 835 Subpart H requires that records be maintained that document compliance with 10 CFR 835. Subpart H requires specific information on the following types of records:

- Individual monitoring records
- Monitoring and workplace records
- Administrative Records

Most of the required radiological records have established retention periods. The retention periods are discussed in DOE Order 200.1, Information Management Program (DOE, 1996a). Individual records may be covered by the Privacy Act; the DOE has codified the Privacy Act in 10 CFR 1008, Records Maintained on Individuals (Privacy Act) (DOE, 1994n).

Detailed guidance on development and maintenance of a radiological exposure recordkeeping and reporting system can be found in DOE Implementation Guide G-10 CFR 835/H1, Rev. 1, Implementation Guides Occupational Radiation Protection Record-keeping and Reporting (DOE, 1994d).

### **3.8 ALARA AND OPTIMIZATION**

The policy of maintaining radiation exposures ALARA has existed in principle since the early 1940s. The evolution of ALARA into a formal program began in the early 1960s. It is well to remember that the ALARA approach was applied to radiation protection far earlier and is much more institutionalized than any comparable approach to other hazards.

Although there is, and has been since the 1940s, a series of official established dose limits, they do not represent ALARA. ALARA is a continuous process of controlling and managing radiation exposure to workers, the general public, and the environment. Although ALARA is based upon protection of people and the environment, the philosophy is also grounded on sound economic and operating principles. The responsibility for maintaining radiation exposures ALARA is not a unique responsibility of management or health physics personnel. It is a responsibility of everyone involved in managing, supervising, or performing radiation work. It is imperative to teach administrative personnel to support the principles and practice of ALARA, and to train all radiation workers to consider ALARA as they prepare for and perform their work.

10 CFR 835 Subpart K “Design and Control” contains specific requirements relating to ALARA considerations for facility design and modification. Also, DOE Order 5400.5 “Radiation Protection of the Public and Environment” (DOE 1990b) contains environmental ALARA requirements.

### **3.8.1 Current Status of ALARA Programs**

Currently, it is common practice in a DOE facility to have a well-structured ALARA plan for the entire facility, with more detailed plans in the various buildings or functional subunits of the facility. There is ordinarily a facility coordinator who administers the overall ALARA plan and reports to top-level management of the facility. Coordinators for the various buildings or subunits of the facility receive guidance from the overall facility coordinator and report the results of their ALARA programs to that individual.

### **3.8.2 Achievement of Goals**

The Radiological Control Manual (DOE, 1994I) provides guidance to contractors (facility) to provide documentation of the ALARA process. To ensure improving radiological performance, at the beginning of each fiscal year, each facility prepares and submits Radiological Performance Goals. At least quarterly, the contractor (facility) provides the contractor senior site executive with an interim status report of the goals. At the end of the calendar year, an Annual Goal Status Report is issued.

Identifying specific ALARA goals in plutonium facilities requires close coordination between the facility ALARA team members (operations, maintenance, and health physics personnel) made up from a cross-section of personnel representing the various work elements of the facility. ALARA goals may be formulated as qualitative or quantitative types of goals, but must be measurable and achievable, with clearly defined endpoints.

### **3.8.3 Quality Assurance**

Important aspects of any ALARA program are the measurement of beneficial effects and the determination that important factors, such as economic impacts, the time involved in accomplishing tasks, and the utilization of personnel, are being optimized. To accomplish these objectives, it is necessary to have a written plan for the ALARA program and high quality records of activities involving exposures to workers, the public, and the environment. These permit comparisons with past experiences and analysis of the recorded activities. In many cases, such studies of the recorded activities not only confirm satisfactory execution of the work, but reveal opportunities for future improvements.

One approach which works very well is the inclusion of an ALARA worksheet along with the RWP. Such a worksheet should be prepared by an individual with responsibilities for the work to be performed, a relatively detailed knowledge of the radiological conditions, and knowledge of what is required to accomplish the task. The worksheet should contain estimates of the time to complete the task and the expected radiation doses that will be received. If any special engineered devices are used to control or reduce personnel exposure, they should be noted on the ALARA worksheet, along with any special instructions that they require. These worksheets provide valuable information for analysis of the effectiveness of the ALARA program for each job.

#### **3.8.4 Technical Aspects**

The technical aspects of ALARA programs include not only the standard equipment regularly used in controlling dose to workers, the public, and the environment, such as facility shielding, ventilation filters, installed and portable radiation measuring instruments, but also many special devices that may be used temporarily. Special devices can be used to provide exposure control and/or containment when it may not be practical without them. These include temporary shields, tents or greenhouses, portable fans, ductwork and filters, and special fixtures to hold highly radioactive materials requiring detailed inspections, repairs, modification, or fabrication. Such devices can permit doing difficult work at low radiation doses, which might not be possible otherwise.

Some of these special devices may have general application and can be kept on hand for use as needed. In some cases, devices would have to be especially fabricated for a specific task. Since this would ordinarily have a significant effect on the cost of doing that job, the economic aspects of doing or not doing the job would have to be carefully evaluated.

#### **3.8.5 Attributes of Effective Review and Audit**

Evaluation of the effectiveness of an ALARA program requires both reviews and auditing. The reviews will include detailed examination of the written ALARA program plan and the records of ALARA activities. The objectives in such reviews are to find if the written plan is being followed, and what is working or not working well. Such reviews can be performed adequately by either a knowledgeable member of the facility staff or an equally knowledgeable outsider. The written report of a review should be directed to a member of management who is responsible for implementation of the ALARA program.

Audits are best performed by an outside health physicist who is sufficiently knowledgeable about work with plutonium and its radiological characteristics that he/she knows where to look for problems and can make appropriate evaluations and recommendations. He should not only examine the ALARA program plan and records, but should also visit the working areas and laboratories in the facility, with a knowledgeable escort who can answer questions about activities and conditions in the facility.

There is nothing really unique in ALARA programs at plutonium facilities, compared with facilities handling other kinds of radioactive materials. However, the radioactivity of plutonium, its potential for criticality, and its relatively high radiotoxicity require somewhat more meticulous surveillance and control than many other radionuclides. Therefore, the detail in ALARA programs for plutonium facilities is likely to be somewhat greater than would be found in ALARA programs for many other facilities.

In any plutonium facility, it is highly desirable to have well-structured ALARA teams in each building or subunit of the facility. Facility goals are to be developed by the facility ALARA teams. Each facility ALARA team chairperson meets with the facility team, and together they develop the calendar-year ALARA goals for their facility. The team chairperson prepares the draft goal report for review by the facility ALARA team and the ALARA Program Office (APO) of the facility. After the draft goals are reviewed, they are returned to the facility team chairperson. The final goal report is then submitted to the facility manager for signature. The signed copy is submitted to the APO to be included in the annual ALARA goal report to DOE. All facility-specific goals should be categorized and reported using the facility-specified format and should include the following:

- **Exposure Reduction.** Goals listed under exposure reduction may reflect occupational or nonoccupational exposure reduction. Exposure to radiological hazards or nonradiological hazards are relevant. Specific jobs for which exposure reduction plans have been developed should be covered in this section. Exposure may be reduced by reducing other hazards that contribute to the difficulty of performing work in radiological areas. For example, reducing noise, reducing heat stress conditions, or improving lighting may facilitate the completion and accuracy of work performed in radiological areas and, thus, reduce exposure. Such opportunities for exposure reduction should be carefully evaluated and appropriate ALARA goals established to make the most of these opportunities.

- **Source Reduction.** Source reduction should concentration on minimizing or eliminating the sources of radiation exposure. Reducing the number of areas with radiological contamination and reducing dose rate are examples of source-reduction goals. Where the presence of nonradiological hazardous materials results in mixed waste, the removal of the hazardous material may have ALARA benefits by reducing the waste classification. Such changes may also reduce exposure at a later time by eliminating the need to store or further treat the waste. In these cases, eliminating the hazardous material may be an appropriate source-reduction ALARA goal.
  
- **Administrative.** Administrative goals typically encompass training, program improvements, procedure revision, or other administrative-type activities. Administrative goals are generally qualitative, so it is difficult to develop endpoints for them. Specific efforts must be made to ensure that adequate closure mechanisms exist for administrative goals.

During all phases of ALARA goal-setting, the facility health physics personnel should be intimately involved in providing advice and expertise on ALARA actions.

When addressing exposure reduction, a cost/benefit analysis should be made to determine the real cost of implementing a dose reduction plan. The Health Physics Manual of Good Practices for Reducing Exposures to Levels that are as Low as Reasonably Achievable (Munson et al., 1988), provides an excellent methodology for conducting a cost/benefit analysis by health physics personnel.

The application of ALARA principles to the performance of work in the field is the main objective of any ALARA program. ALARA design, engineering, planning, and administration come to fruition in maintaining exposures ALARA to workers and the public. The operational application of ALARA requires cooperation and coordination of many functional groups, including radiation protection, operations, maintenance, planning and scheduling, training, engineering, and administration.

The primary responsibility for controlling radiation exposure during operations rests with the individual and his/her immediate supervisor. The support functions provide the training, resources, guidance, and measurements, but it is in the application that the effectiveness of an ALARA program is realized. Operational measures for controlling exposure must be

applied to assure that any work with radioactive materials is carried out in the safest manner reasonable. Both engineered and administrative control measures should be used for limiting exposure.

Engineered controls should be utilized whenever possible. In addition, periodic verification of the continued effectiveness of these controls should be performed by facility health physics personnel. Ventilation and filtration systems should be routinely checked and inspected to assure that operation is maintained within the design criteria. The integrity of shielding, the reliability of equipment, and the calibration of instruments should likewise be routinely verified.

Although administrative controls are not an adequate substitute for engineered features, they are necessary. They are a part of the management systems developed and implemented to provide guidance, direction control, and limitations for activities. Administrative controls include the documents that describe organizational interfaces and prescribe controls for radiation protection. Administrative controls, especially procedures, should be reviewed by those responsible for ALARA to ensure that radiation exposure activities include dose limitation considerations.

Factors that must always be considered in an ALARA program are the costs and benefits. This is especially important when the identified benefit represents a very small increment of radiation dose reduction. Funds for dose reduction should always be applied to actions which will achieve the greatest dose reduction for the cost.

The final decontamination and decommissioning (D&D) of a plutonium facility should be given consideration in both the original design of the facility and any modifications done to the facility during its operating lifetime. Likewise, D&D should be given consideration in choosing operating processes and practices for the facility, including any changes in processes and practices during its operating lifetime. Both design and operating activities can affect the radiation levels and personnel doses encountered by workers who perform the D&D activities. To the extent practicable, design and operations should provide for radiation levels that are ALARA during D&D activities.

The successful implementation of an ALARA program requires the commitment, support, attention, and efforts of all members of an organization. In facilities in which the radiation exposures are already relatively low, implementation of the ALARA concept is particularly



challenging. The reduction of radiation doses to ALARA levels demonstrates to workers and the public a continued emphasis, commitment, and concern for health and safety.

### 3.9 CONDUCT OF OPERATIONS

The organization and administration of operations should ensure that a high level of performance in DOE facility operations is achieved through effective implementation and control activities.

Administration of operations activities should recognize that protection of the environment, maintaining high-quality safety, and productivity are compatible goals. The DOE policies should describe the standards of excellence under which the facility is expected to operate. Clear lines of responsibility for normal and emergency conditions must be established. Effective implementation and control of operating activities are achieved primarily by having readily accessible written standards for operations, periodical monitoring and assessment of performance, and personnel accountability for performance. For a more detailed discussion, see DOE Order 5480.19, Conduct of Operations Requirements for DOE Facilities (DOE, 1990a).

A high level of performance in DOE operations is accomplished by management establishing high operating standards and then by communicating the operating standards to workers:

- By providing sufficient resources to the operations department
- by ensuring that personnel are well trained by closely monitoring performance in operations
- by holding workers and their supervisors accountable for their performance in conducting activities.

Senior management establishes operating standards, considering input from workers when appropriate. Working-level personnel will more strongly support the standards when they have had appropriate input into their development. The standards should define operating objectives, establish expected performance levels, and clearly define responsibilities in plant operations. Standards for operating activities should be integrated into operations department procedures and programs. Operating standards should also be communicated to workers by training them in operating practices and by having supervisors monitor and guide work involving facility operations. Sufficient staff, facilities, equipment, and funding should be allocated to permit the operations department to effectively perform its functions. Performance in operations should be closely monitored by facility management, preferably using operating reports and goals, so that the

performance of the operations department can be effectively measured. Operations personnel should be held accountable for their performance through supervisor counseling, performance appraisals, and, when necessary, disciplinary measures. Remedial training should be provided when appropriate.

The health physics organization, as a support element, must ensure that all aspects of radiation safety are considered in the establishment of operations standards and policy. A well instituted cooperative relationship between operations and health physics is paramount to the health and safety of workers and the public and to protection of the environment.

A plutonium facility should have a written policy on radiation protection, including an ALARA policy. All radiation protection procedures and controls should have recognizable or formal technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to assure that all copies are accounted for and that all new procedures are included in the historical files. A designated period of time for holding the historical files should be established. DOE Order 200.1 (DOE, 1996a) and ANSI N13.6-1989 (ANSI, 1966) provide guidance on how long to keep historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur.

The radiation protection procedure system should provide for but not be limited to, the following topics: radiation work procedures, posting and labeling, instrument calibration, and provision for audits.

### **3.9.1 Radiation Work Procedures**

Radiation work procedures, including RWPs, survey procedures, ALARA reviews, sample counting, and other task procedures, fall within the requirements for conduct of operations. All sections of DOE Order 5480.19 (DOE, 1990a) apply. The guidance and requirements of Section XVI, "Operations Procedures," is especially pertinent to radiation work procedures. Procedures are a key factor affecting radiation protection performance. Appropriate attention should be given to writing, reviewing, approving, and monitoring implementation of radiation protection procedures. There should be documented qualification and training requirements for those who prepare and approve procedures. A formal approval process

should be established. Procedure changes and revisions should be subject to the same review and approval process as the initial procedure.

Personnel should be trained in the use of the procedures they will be expected to perform. For RWPs, workers should read the RWP and verify by signature that they have read it, understand its contents, and will comply with its requirements in the conduct of the work. Procedures should be available for personnel use. The RWPs should be posted at the entrance to the work location. There should be a system in place to assure that posted copies of all work procedures, including RWPs, are current.

### **3.9.2 Posting and Labeling**

The requirements for posting and labeling of working areas because of the presence, or potential presence, of radiation and/or radioactive material are specified in 10 CFR 835, Subpart G (DOE, 1993c). Guidance in implementing the regulatory requirements can be found in G-10 CFR 835/G1-Rev. 1, Implementation Guide. Posting and Labeling for Radiological Control (DOE, 1994e), and the Radiological Control Manual (DOE, 1994l). Conformance of conduct of operations requirements should assure a reasonable degree of uniformity in the posting and the signs used, as well as verifying that operator aids and other posted information do not interfere with necessary radiological posting. It is necessary to formally review posting of radiological areas in the same manner that the posting of operating aids is reviewed, in conformance with DOE Order 5480.19 (DOE, 1990a).

### **3.9.3 Calibration of Instruments**

The status of installed and portable radiological instruments should be well known and appropriate to the use. (Calibration of radiological instruments is discussed in Section 3.5.2.)

”Ownership” of installed radiological dose rate and airborne contamination monitoring instrumentation should be well known and the responsibility and authority for calibration, repair, and notification clearly established. Because such information is often used by more than one group, formal notification procedures should be established to cover those times when the instrument is out of service or beyond the required calibration schedule. Configuration control and quality assurance requirements for installed systems should be established commensurate with their safety significance.

For portable instrumentation, conduct of operations requirements are normally built into the routine calibration and survey program. Functional checks are routinely made to verify calibration, instruments are checked to assure that they are within the calibration period, and survey procedures require identification of the instruments used so that if a problem is later found, measurements can be repeated.

### **3.9.4 Audits**

Conduct of operations does not, in itself, contain requirements on auditing. Inspections, audits, reviews, investigations, and self-assessments are part of the checks and balances needed in an operating program. Auditing is one of the many tools that line management has at its disposal to identify problems. Regulation 10 CFR 835.102 requires internal audits of all functional elements of the radiation protection program no less frequently than every 3 years. These audits are to include program content and implementation.. Each one of the 18 topics addressed in DOE Order 5480.19 (DOE, 1990a) should be subject to both internal self-assessment and external auditing to assure effective implementation of their requirements. Any deficiencies identified should be documented and corrective actions aggressively pursued and tracked to completion. The self-assessment and audit process should include conducting trend analyses and root cause evaluations of deficiencies and communication of results throughout the organization.

### **3.9.5 Decommissioning of Weapons and Weapon Facilities**

Decommissioning of nuclear weapons and nuclear facilities is subject to the same conduct of operations requirements as operating facilities. In general, some components, once they are separated, can be downgraded in safety significance. Also, facilities undergoing decommissioning will have fewer safety systems.

During decommissioning, status control and shift turnover are extremely important considerations and must be done in accordance with DOE Order 5480.19 (DOE, 1990a). Posting and labeling of radiological areas are also an increasing challenge because of the rapidly changing radiological status. In extreme cases, it may be desirable to have workers review or sign the RWP each day to ensure they are aware of the status.

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## **4.0 CONTAMINATION CONTROL**

The primary control for contamination in a plutonium plant is the facility design. Contamination is confined primarily by enclosing the process areas and using controlled ventilation systems. Appendix C of this manual addresses the different levels of confinement in a plutonium facility. The design objective for the confinement system is to essentially prevent or minimize exposure of plant personnel and the public to airborne contamination [DOE Order 6430.1A (DOE, 1989b)]. To ensure that this objective is met, additional attention should be given to air contamination control, surface contamination control, and personnel contamination control. Radiological controls for the workplace should ensure that radionuclides are contained and handled properly and that intakes, if they occur at all, are negligible to the extent achievable with state-of-the-art technology. However, much of the current effort involves decommissioning of no-longer-needed production facilities. The lack of engineered controls or the systematic removal of existing controls during the decommissioning process introduces a completely different set of circumstances that requires special attention for adequate contamination control and worker and public protection.

### **4.1 AIR CONTAMINATION CONTROL**

To achieve the design objective of preventing (or at least minimizing) internal exposure of plant personnel, airborne contamination must be confined to process enclosures which have adequate air cleaning systems. Because both equipment and personnel errors can compromise designed protection and because older facilities may already have unconfined plutonium, air monitoring and other contamination control measures are needed. Experience has shown that the most common route for inadvertent plutonium deposition in man is by inhalation even though intakes may also occur by accidental ingestion or by wound contamination. In facilities being decommissioned, the use of temporary containment structures, interim ventilation systems, and administrative controls such as protective clothing and respirators may be required to replace engineered systems.

10 CFR 835.1002 requires that for the control of airborne radioactive material, the design objective shall be, under normal conditions, to avoid releases to the workplace atmosphere and in any situation, to control the inhalation of such material by workers to levels that are ALARA; confinement and ventilation shall normally be used. See Appendix C for Facility Design guidance.

Note: The use of ventilation systems may require the approval of Facility Criticality Safety personnel because these systems may concentrate fissionable material.

#### **4.1.1 Internal Versus External Dose Philosophy**

The overall goal of radiological protection is to minimize the total dose to the individual. However, because of the difficulties and cost of an adequate internal dosimetry program for plutonium exposure, it is best to avoid all internal exposures during routine operations and anticipated abnormal events by facility design features and personnel protective equipment. As stated above, this is an extremely challenging goal for those facilities in environmental remediation. The conditions encountered in decommissioning and environmental restoration will typically place a heavy reliance on administrative controls.

#### **4.1.2 Purpose of Air Monitoring**

Airborne contamination surveys are performed for the following reasons:

- Prompt detection of airborne contaminants for worker protection
- Personnel exposure assessment
- Monitoring of trends within the workplace
- Special studies.

Of primary importance is the prompt detection of airborne contaminants. The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the person to intercept the airborne materials before they reach the individual. With the numerous sources and mobility of the workers, interception under all conditions is difficult, if not impossible, to achieve. To aid in early detection samples of airborne materials should be taken as close to their points of origin as practicable to maximize the probability of their detection (airborne concentrations are at a maximum at their points of origin). Detailed guidance for the placement of air samplers and monitors, selection of system characteristics and requirements, and maintenance and calibration of the equipment is available in the Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) and Air Sampling in the Workplace (NRC, 1993).



#### **4.1.3 Regulations and Limits**

The regulations for control of radiation work are covered in 10 CFR 835 (DOE, 1993c). Additional requirements and guidance for implementation is provided in the DOE Radiological Control Manual (DOE, 1994I), and the Implementation Guides. While many of the topics included in the Implementation Guides relate to plutonium contamination control, specific guidance on contamination control has not been provided. The limits established for plutonium and other transuranic elements for contamination areas, high contamination areas, and airborne radioactivity areas are given in 10 CFR 835.603 and Appendix D of 10 CFR 835 and are summarized in Table 4.1.

#### **4.1.4 Uncertainties and Limitations**

Because plutonium is relatively difficult to detect and quantify, it is important to consider the uncertainty in the measurements when designing a plutonium monitoring program. Although the design objective of the facility will likely be no airborne plutonium contamination, the reality will be a measurement that ensures airborne plutonium is below an acceptable lower limit of detection. The sampling and monitoring program will need to be designed not only for prompt detection of airborne contamination, but to assure that samples are representative of the air that the workers are breathing and have a low enough limit of detection that only negligible doses could go undetected. Bioassay and in vivo analysis of plutonium should confirm the negligible exposure that the air monitoring program documents and provide a baseline for any accident- or incident-related exposures. The need for an effective sampling and monitoring program is even more critical in the rapidly changing environment of decommissioning activities.

Numerous factors enter into any determination of plutonium contamination levels and the risk to workers. Some of these factors are detection efficiency of the measuring instrument, collection efficiency of the smear media or air sample filter, the location of the smear or air sample in relation to the source of contamination, the physical and chemical properties of the contamination, the representativeness of the air sample to the air being breathed by the worker, the engineered controls available, and the protective equipment used. All of these factors must be considered in the development of a plutonium contamination control program and in evaluating the actions required for personnel protection.

**Table 4.1.** Surface Radioactivity Values,<sup>(1)</sup> dpm/100 cm<sup>2</sup>

Nuclide	Removable <sup>(2),(4)</sup>	Total (Fixed+Removable) <sup>(2),(3)</sup>
U-nat, <sup>235</sup> U, <sup>238</sup> U, and associated decay products	1000	5000
Transuranics, <sup>226</sup> Ra, <sup>228</sup> Ra, <sup>230</sup> Th, <sup>228</sup> Th, <sup>231</sup> Pa, <sup>227</sup> Ac, <sup>125</sup> I, <sup>129</sup> I	20	500
Th-nat, <sup>232</sup> Th, <sup>90</sup> Sr, <sup>223</sup> Ra, <sup>224</sup> Ra, <sup>232</sup> U, <sup>126</sup> I, <sup>131</sup> I, <sup>133</sup> I	200	1000
Beta-gamma-emitters (nuclides with decay modes other than alpha emission or spontaneous fission) except <sup>90</sup> Sr and others noted above <sup>(5)</sup>	1000	5000
Tritium organic compounds; surfaces contaminated by HT, HTO, and metal tritide aerosols	[Reserved]	[Reserved]

<sup>(1)</sup> The values in this table apply to radioactive contamination deposited on, but not incorporated into the interior of, the contaminated item. Where surface contamination by both alpha- and beta-gamma-emitting nuclides exists, the limits established for alpha- and beta-gamma-emitting nuclides should apply independently.

<sup>(2)</sup> As used in this table, dpm (disintegrations per minute) means the rate of emission by radioactive material as determined by correcting the counts per minute observed by an appropriate detector for background, efficiency, and geometric factors associated with the instrumentation.

<sup>(3)</sup> The levels may be averaged over 1 m<sup>2</sup> provided the maximum surface activity in any area of 100 cm<sup>2</sup> is less than three times the value specified. For purposes of averaging, any square meter of surface shall be considered to be above the surface radioactivity values if: (1) From measurements of a representative number n of sections it is determined that  $1/n \sum S_i \geq$  surface radioactivity values where  $S_i$  is dpm/100 cm<sup>2</sup> determined from measurement of section  $i$ ; or (2) it is determined that the sum of the activity of all isolated spots or particles in any 100 cm<sup>2</sup> area exceeds 3G.

<sup>(4)</sup> The amount of removable radioactive material per 100 cm<sup>2</sup> of surface area should be determined by swiping the area with a dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency. (Note--The use of dry material may not be appropriate for tritium.) When removable contamination on objects of surface area less than 100 cm<sup>2</sup> is determined, the activity per unit area should be based on the actual area and the entire surface should be wiped. Except for transuranics and <sup>228</sup>Ra, <sup>227</sup>Ac, <sup>228</sup>Th, <sup>230</sup>Th, <sup>231</sup>Pa and alpha-emitters, it is not necessary to use swiping techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination.

<sup>(5)</sup> This category of radionuclides includes mixed fission products, including the <sup>90</sup>Sr which is present in them. It does not apply to <sup>90</sup>Sr which has been separated from the other fission products or mixtures where the <sup>90</sup>Sr has been enriched.

#### 4.1.5 Samples and Instrumentation

For plutonium facilities, both air sampling and air monitoring are essential elements of the radiological control program. Real-time air monitoring using alpha-sensitive continuous air monitors (CAMs) should be used to alert workers to rapid degradation of radiological conditions. The air sampling system with a lower limit of detection must be adequate to provide continuing assurance that personnel exposures are within limits and ALARA.

The characteristics of a good plutonium CAM include

- A lower limit of detection equal to or better than 8-DAC-h
- high reliability with a minimum of spurious alarms
- a stable and constant flow air mover
- stable and documented detector efficiency with geometry, filter collection efficiency, self-attenuation, etc., considered
- methodology for radiation discrimination and natural radioactivity discrimination
- system for activating an alarm
- shielding for extraneous sources of interference such as radiation, radiofrequency, temperature, and vibration
- mechanical and electrical ruggedness
- ease of maintenance and calibration.

A plutonium air sampling program typically includes a system of fixed head air samplers to quantify air concentrations in the workplace. The basic characteristics of the sampling equipment remain the same except that there is normally less flexibility in locating the sampling heads but more flexibility in selecting and operating the counting instrumentation. In many instances, installed sampling systems may no longer be operational or may be in the wrong locations. In those instances, portable air sampling systems, both impactor-head type or filter type may be used to provide required worker protection.

#### **4.1.6 Sample Analysis**

Plutonium air samples are typically analyzed by alpha counting, alpha spectral analysis, or chemical analysis. The technique used will depend upon the filter media used, the physical and chemical state of the contaminate, the urgency for the data, interfering radionuclides, and other factors. Authoritative guidance in establishing plutonium air sampling counting and analysis methods can be found in NCRP Report No. 58, A

Handbook of Radioactivity Measurements Procedures (NCRP, 1985) and in Air Sampling in the Workplace (NRC, 1993).

#### **4.1.7 Monitoring Strategies and Protocols**

The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the potentially exposed worker (or member of the public) to intercept the airborne materials before they reach the individual. With the numerous sources and mobility of the workers, interception under all conditions is difficult, if not impossible, to achieve. Samples of airborne materials should be taken as close to their points of origin as practicable to maximize the probability of their detection (airborne concentrations are at a maximum at their points of origin).

Fixed probes that are positioned to intercept releases from recognized major potential sources should be used along with portable air samplers for planned activities with known potentials for airborne release of contaminants and for temporary storage of contaminated materials in areas of low air flow. If the workplace exhaust system can be shown to provide rapid, essentially quantitative clearance of airborne contamination, fixed probes that sample the exhaust system may be adequate for routine coverage of unplanned activities. If justified by documented studies, other sampling arrangements may be used that provide improved "total" coverage of the workplace environment for the early detection of airborne contamination.

Those responsible for the rapid and reliable detection of airborne plutonium should consider the following workplace characteristics in evaluating monitoring systems and working environments (Mishima et al., 1988):

- The airflow patterns and airborne transport of plutonium in the workplace
- the location of personnel within the workplace during various processing procedures
- the location at which the airborne plutonium sample should be intercepted before the sample is inhaled by workers

- the ability of the system to transport an undistorted sample to the collection media or measurement device
- the collection and retention efficiency of the collection medium
- the efficiency of the measurement device in measuring the plutonium collected and differentiating the plutonium from other materials present
- the accuracy and reliability of the system.

Guidance for each area listed above is provided in Mishima et al. (1988).

## **4.2 SURFACE CONTAMINATION CONTROL**

Controlling plutonium surface contamination is essential because it may easily be resuspended in air and/or transferred to other surfaces. The following elements are important for controlling surface contamination: keeping plant surfaces clean; monitoring, reporting, and tracking contamination levels; and establishing appropriate control zones with limits and action levels for those zones.

### **4.2.1 Plant Surfaces**

Good housekeeping practices are essential in keeping plant surfaces clean. Periodic housekeeping should be performed within contaminated areas to minimize the buildup of contamination and contaminated waste. Periodic decontamination both within contaminated glove boxes and in the general work area should be conducted to minimize removable contamination.

In some instances, it may be appropriate to apply fixatives to minimize the movement of plutonium contamination. However, it is generally desirable to attempt decontamination first. If decontaminating is not successful or perhaps, not appropriate for the job scope, a fixative may be appropriate. If a fixative is used, typically a paint, two layers of fixative should be used, with the bottom coat yellow and the top coat a different color. When the yellow begins to show through the top coat, additional fixative should be applied. Also, for areas which have had a fixative applied over plutonium contamination, a routine

contamination survey should be conducted to assure that no contamination has become movable over time.

In some cases a strippable coating may be used to allow easy decontamination at the completion of a job. These strippable coatings are sometimes used to decontaminate areas. An aerosol fixative is also available that can be pumped into a room, glovebox, or other work space, that coats all exposed surfaces, including the underside of components. This allows work to proceed without disturbing contamination.

Note: The use of fixatives may require the approval of Facility Criticality Safety personnel because fixatives may concentrate or moderate fissionable material.

Outside areas may also require a fixative to minimize the spread of contamination. Historically, some outside contaminated areas have been covered with asphalt to fix contamination. This is not a desirable material to use because it creates a mixed hazardous waste as well as significantly increasing the volume of contaminated material for disposal. Two substances that currently are used as an interim fixative for outside soil/surface contamination areas are 1) a derivative of pine tar (toll oil), which forms a non-toxic surface fixative that is hard and appears to have a relatively durable surface and 2) a mixture of white glue and water (enduro seal), which is easily sprayed on and sets rapidly to a firm surface. A water to glue ratio of about 25 to 1 appears to perform well in preliminary tests. Both of these fixatives are only interim measures because of eventual degradation from the elements. For more localized areas where a permanent fixative/cover is needed, a sprayable concrete (Shotcrete) is available. A disadvantage of this material is cracking, which defeats the sealing surface. Another material that can be used as a carpeting for outside contamination is a spray-on two-part polymer that provides a flexible, semidurable cover. The characteristics of the cover can be adjusted to vary water transmission and the color can be changed to inhibit growth under the covering. The major problem for outside use of all of these fixatives is the invasion and actions of biota. Mice, rabbits, other wildlife as well as plant growth tend to burrow under any covering and spread the contamination. While these measures do not permanently solve the problem, they may provide a method of preventing the spread of contamination until a permanent, acceptable solution is determined.

**4.2.1.1 Housekeeping**

The three housekeeping practices listed below should be followed in a plutonium facility as part of the Conduct of Operations [see DOE Order 5480.19 (DOE, 1990a)]:

- The inventory of contaminated and potentially contaminated scrap and equipment should be kept to a minimum because all such materials are subject to special monitoring and accountability.
- Radioactive contamination should be controlled and the spread of contaminants and the potential for accidents involving contaminants shall be minimized. (In at least one instance, poor housekeeping contributed to a serious criticality accident.) Management at all levels should continuously emphasize the importance of good housekeeping, and operating procedures should be written to ensure good housekeeping practices.
- Measures shall be taken to maintain radiation exposure in controlled areas as low as is reasonably achievable through facility and equipment design and administrative control (10 CFR 835.1001).

Where possible, materials that are not absolutely necessary to an operation should be kept out of the contaminated or potentially contaminated area. It is very important to minimize the creation of TRU waste. All packaging and unnecessary protective coverings should be removed before materials are introduced into the process area. Likewise, items that are not necessary to the process should be promptly removed, particularly from glove boxes, and not left to accumulate and become safety hazards, potential fire hazards, sources of radioactive (dust) accumulation, or sources of exposure.

Good housekeeping practices inside glove boxes should emphasize fire and explosion control. Only metal or nonflammable plastic containers should be used for the accumulation of scrap and wastes of any kind in the glove boxes and throughout plutonium facilities. Accumulation of combustible materials in glove boxes should be minimized. When explosive, flammable, or volatile liquids are

allowed, they should be rigidly controlled and used only in inert gas atmospheres unless a safety analysis review shows it is safe to do otherwise. All residues should be removed immediately at the conclusion of each job or cleaning operation.

Considerable effort has been expended on the development of coated and corrosion-resistant tools. Some efforts have been marginally successful, but in most cases throw-away tools are favored. Electropolishing of contaminated metal tools and equipment has been shown to be a good method of decontamination and allows for their reuse in some cases or disposal as noncontaminated waste. Where possible, all tools with sharp edges or points (e.g., screwdrivers, ice picks, scissors) should be kept out of glove boxes.

Management should constantly demand good housekeeping. Mandatory, routine clean-up periods are becoming more common due to the increasing cost of storing and disposing of contaminated materials. Better housekeeping is required due to real-time, computerized accountability for nuclear materials. It has been demonstrated that kilogram quantities of plutonium oxide dust can accumulate in glove boxes unless they are routinely cleaned. Much of the exposure to workers originates from layers of plutonium oxide dust on the surface of gloves and the internal surfaces of glove boxes. In processes where plutonium oxide powder is handled, the glove boxes should be cleaned weekly to reduce the accumulation of dust layers and to reduce worker exposure. Although difficult to achieve and maintain, good housekeeping is equally essential during decommissioning of plutonium facilities.

#### **4.2.1.2 Vacuuming**

The subject of vacuuming within a glove box is somewhat complex. Experience has shown vacuuming to be the most effective and quickest way to clean a controlled-atmosphere (dry) glove box. It is not particularly effective for high-humidity or wet-process glove boxes, particularly those that involve acids. After acids have been used in a glove box, washing and wiping is the preferred method of cleaning the etched surfaces.



Vacuuming is particularly effective in dry-atmosphere and inerted enclosures where the levels of radioactive dust can quickly increase personnel exposure. In many cases, vacuuming reduces the exposure level more than a wipe down with a damp cloth, and it can be done more quickly and with less waste material generated. Two factors weigh against vacuuming: possible safety hazards from electrical sparks, and the occasional difficulty of operating in inert atmospheres (although the last item need not be of importance). However, in dry glove boxes with dusty operations using high-exposure plutonium, personnel exposure control is a problem and vacuuming is a quick and effective method of keeping the dust and exposure rates under control and should be considered.

Note: The use of vacuum cleaners may require review by Facility Criticality Safety personnel because vacuum cleaners are likely to concentrate fissionable material.

The use of vacuum cleaners for contamination control requires careful consideration and strict controls to assure that the process does not spread contamination. As a minimum, all vacuums used for radioactive material should have HEPA filtration on the exhaust. In some instances, the additional precaution of having the exhaust vented into a process ventilation system should be considered.

#### **4.2.2 Reporting and Documenting Contamination Levels**

Radiological control programs require the performance of contamination surveys to determine existing conditions in a given location. Maps with sufficient detail to permit identification of original survey locations should be maintained. Records should contain sufficient detail to be meaningful even after the originator is no longer available. Contamination surveys should be recorded on appropriate standard forms and include the following common elements:

- Date, time, and purpose of the survey
- General and specific location of the survey
- Name and signature of the surveyor and analyst

- Pertinent information needed to interpret the survey results
- Reference to a specific Radiological Work Permit if the survey is performed to support the permit [see DOE Radiological Control Manual, part 751.1 (DOE, 1994I)].

Records should be maintained to document changes in monitoring equipment, techniques and procedures [see DOE Radiological Control Manual, part 751.2 (DOE, 1994I)].

In addition, records of contamination surveys should include, at a minimum, the following information:

- Model and serial number of counting equipment
- Contamination levels (using appropriate units) and appropriate supporting parameters, including counting efficiency, counting time, correction factors, type of radiation, and whether the contamination was fixed or removable
- Location of areas found to contain hot particles or high concentrations of localized contamination
- Follow-up survey results for decontamination processes cross-referenced to the original survey (see DOE Radiological Control Manual, part 754).

Records for the release of material and equipment from radiological areas to controlled areas shall describe the property, the date on which the release survey was performed, the identity of the individual who performed the survey, the type and identification number of the survey instrument used, and the results of the survey (10 CFR 835.1101(d)). Additional details on radiation records can be obtained from the Implementation Guide. Occupational Radiation Protection Record-keeping and Reporting (DOE, 1994d).

All skin and personal property contaminations should be documented and evaluated to help improve the contamination control program. Documentation should include the following:

- The person's name and work group

- The location, amount, and type of skin or personal property contamination
- The results of decontamination
- A description of circumstances involved in the occurrence, such as radiation work permit number, protective clothing required, and protective clothing actually used.

#### **4.2.3 Characteristics of Plutonium Contamination**

There are few characteristics of plutonium contamination that are unique. Plutonium contamination may be in many physical and chemical forms. (See Section 2.0 for the many potential sources of plutonium contamination from combustion products of a plutonium fire to radiolytic products from long-term storage.) The one characteristic that many believe is unique to plutonium is its ability to migrate with no apparent motive force. Whether from alpha recoil or some other mechanism, plutonium contamination, if not contained or removed, will spread relatively rapidly throughout an area.

#### **4.2.4 Monitoring**

Radiation workers are often assigned tasks that conceivably could expose them to radioactive material. It is not sufficient to rely exclusively on equipment design to minimize contamination and exposure in the workplace. A radiation protection program shall include both monitoring of the workers (discussed in Section 4.3) and monitoring of the conditions in the workplace (10 CFR 835 Subpart E). Both functions are essential to a good radiation monitoring program.

Continuous radiation monitoring should be provided during the periods of high or unusual risk associated with the work in the area. Periods of high or unusual risk include the potential or actual breaching of the integrity of the glove-box or associated systems, including such maintenance as replacement of panels, glove changes, bag-out operations, replacement of filters, or repair of vacuum systems. Work that involves the use of temporary enclosures (greenhouses or glovebags) may also be provided with continuous coverage by an RCT, if the hazard is sufficient to warrant such measures. For decommissioning, most activities will be new, unique, and have no historical precedent.

Consequently, high and unusual risks may become the norm and the use of temporary controls and continuous coverage the routine.

Monitoring of the workplace is an essential element of every routine surveillance program. It can be effectively accomplished using any or all of the techniques that are discussed in this section. The rigor with which all of the various elements of a radiation monitoring program are applied should be tailored to meet the needs of the individual work areas and should depend on the kind and quantity of radioactive material present and its potential for dispersion. Each program should be designed to meet existing needs, but also should be flexible to allow for incorporation of the possible advantages to be provided by the various available monitoring practices. Monitoring practices include, but are not limited, to the following:

- Contamination surveys of the workplace
- Release surveys
- External exposure surveys
- Airborne contamination surveys
- Routine surveillance by an RCT.

#### **4.2.4.1 Contamination Surveys of the Workplace**

The radiation monitoring program should include documented survey procedures, a system for maintaining survey results, and contamination control limits for “fixed” and “removable” contamination. The results of contamination surveys should be reported in activity per area (e.g., dpm/100 cm<sup>2</sup>) except for large-area swipes and swipes of very small items. This permits interpretation of the recorded data without requiring knowledge of instrument efficiency or geometry.

All workplaces should be monitored for contamination levels on a regularly scheduled basis. The frequency of such surveys will depend on the potential for dispersion of the radioactive material. As a minimum, all gloves, work surfaces,

floors, equipment, etc., within the workplace should be surveyed according to the frequencies listed in the DOE Radiological Control Manual (1994l).

The change room and other support facilities within the controlled area should be surveyed for contamination daily. Continuous air monitors, survey instruments at step-off pads, and hand and shoe counters should be functionally tested daily or once per shift in support of the weekly and monthly surveys. These frequent surveys are also part of the routine surveillance program and permit immediate follow-up if low-level contamination is detected to minimize the potential for major incidents. Some fixtures and support areas outside the controlled area, such as door knobs and telephones of adjacent offices and the lunchroom, should also be surveyed daily. Other support areas should be surveyed monthly. If routine survey results detect any contamination in a given area, more detailed surveys must be performed to determine the extent of the contamination. An investigation should be initiated to determine the source of the contamination and the cause.

To preclude the possibility that contaminated waste would be disposed of as ordinary waste, 1) all process and controlled area waste should be considered contaminated, and 2) mechanisms should be established that prevent the mixing of contaminated and noncontaminated waste.

#### **4.2.4.2 Release Surveys**

For transuranic radionuclides, the contamination level (fixed and removable) at which surfaces are considered contaminated are listed in Appendix D of 10 CFR 835 (DOE 1993c). That document also specifies the criteria for the release of materials and equipment to Controlled Areas.

This document concerns release to controlled areas only. The detailed requirements for unrestricted release of materials and equipment are found in DOE Order 5400.5 (DOE, 1990b). Refer to that document for guidance regarding unrestricted releases.

#### **4.2.4.3 External Exposure Surveys**

To delineate the levels involved, measurements of external exposure should be made at the time a program is established at all locations where personnel exposure occurs. Additional photon and neutron measurements should be made at the same frequency as the contamination surveys. The buildup of plutonium contamination in glove boxes and on gloves and equipment may contribute substantially to the external dose rates.

#### **4.2.4.4 Measurement and Survey Techniques**

This section discusses four types of contamination surveys that are typically used in DOE facilities. Surveys for removable contamination include a large-area wipe survey and a technical swipe or smear survey. Surveys for total/fixed contamination include a scan survey and a statistically based survey. These surveys, or a combination of them, are used to survey material for release from radiological control. The appropriate use of each type of survey is discussed.

##### **Surveys for Removable Contamination**

Two types of surveys are used for removable contamination: a large-area wipe survey and a technical swipe or smear survey.

A large-area wipe survey is used to detect gross removable contamination. A large-area wipe survey is typically performed using a large floor cloth and a dust mop type handle to wipe large areas. This technique tends to concentrate any low levels of removable contamination that may be present. The surface to be wiped and the wiping material should be industrially clean (i.e., free of debris, grease, etc.) to reduce self-absorption of alpha contamination. The survey is performed by wiping the surface of the area being surveyed and conducting frequent checks of the cloth using a portable instrument. For detection of alpha-emitting isotopes, a nonabsorbent material should be used. Removable contamination will be accumulated and concentrated on the wipe, increasing the probability of its detection. Checking for contamination is conducted by placing an alpha-measurement instrument approximately 0.25 in. (0.6 cm) from the surface of the wipe for 5 seconds, and the count rate observed. If there is no increase above background, then the wipe may be placed in contact with the detector. If no radioactivity above background is measured, then the material is not contaminated with removable contamination. If radioactivity above background is measured, the material is contaminated. Depending upon the specific circumstances, a series of technical smears may be required to locate and quantify the contamination within the area covered by the large-area wipe. In most instances, if contamination is detected on the large-area wipe, decontamination should be considered.

For transuranic radionuclides, the guideline values for removable contamination are lower than the MDA of portable instruments. During a wipe survey, the surface area of the material must be large enough that the quantity of radioactivity collected on the swipe will be greater than the MDA of the instrument. Wipe surveys of areas smaller than this minimum surface area require more sophisticated measuring instruments, such as a scaler measurement, and the entire surface of the material should be wiped. The minimum area for using a large-area wipe survey is given by

$$A_{\text{minimum}} = \frac{\text{MDA}}{L} \times 100 \text{ cm}^2 \quad (4.1)$$

where L is the removable surface radioactivity value in dpm/100 cm<sup>2</sup> of the potential contaminant, given in Table 4.1. and MDA is in dpm.

The purpose of a technical smear survey is to locate and quantify removable contamination that is known or suspected to exist. For small items, a technical smear may be used at any time to verify the item's contamination status. A technical smear or swipe survey is performed by wiping a cloth, paper, plastic foam, or fiberglass disk over a 100-cm<sup>2</sup> area of the surface. The wipe should be taken with a dry medium using moderate pressure. A common field practice is to use two fingers to press the wipe medium against the surface to be wiped. The wipe is then moved along an "S" shaped path that has a nominal length of 8 in. (20 cm) to 10 in. (25 cm).

When the potential contaminant emits alpha radiation, paper or fiberglass filter papers should be used to assure that alpha activity is not attenuated by becoming imbedded in the wipe. To improve the detection limit, smears may be taken over areas larger than 100 cm<sup>2</sup>. However, the size of the area smeared should be limited to prevent buildup of material (radioactive or otherwise) that would attenuate alpha radiation. The current practice at DOE facilities is to use the 100-cm<sup>2</sup> area as the minimum size of objects being smeared. Appropriate corrections should be made for objects smaller than 100 cm<sup>2</sup>.

If contamination is detected during a scan survey for fixed contamination, a survey for removable contamination should be performed to determine if the contamination is fixed and to quantify any removable contamination. The survey should be performed using a small piece of absorbent material, such as a standard paper smear. This type of survey for removable contamination is often called a technical smear survey. If no contamination above the guideline values for removable contamination in Table 4.1 is detected during the smear survey, the contamination is fixed, and the area should be posted appropriately.

A technical smear survey may be used routinely to detect removable contamination, especially for contamination surveys of radiological areas.



**Scan Survey for Fixed Contamination**

A scan survey for fixed contamination requires passing a portable instrument over the surface of the area being surveyed at a fixed, known scan speed and at a specified distance from the surface. Typically, the scan speed is 2 in./s (5 cm/s) and the maximum distance is 0.25 in. (0.6 cm) for alpha contamination instruments, but this can vary depending on the instrument, probe configuration and background. A scan survey should be used to survey material that resides in an area controlled for contamination purposes, an area where unsealed radioactive sources are used, or a radiological buffer area surrounding an area controlled for contamination purposes. A scan survey in conjunction with a wipe survey should be used to release from radiological control material with a total surface area less than 5 ft<sup>2</sup> (0.46 m<sup>2</sup>). A statistically based survey, which will be discussed later, should be used to release from radiological control material with a surface area greater than 5 ft<sup>2</sup> (0.46 m<sup>2</sup>).

During the performance of scan surveys, the audible response of the instrument is faster than the needle deflection. Therefore, audible response should be used in conjunction with meter readings. For alpha surveys, the surveyor should pause for 3 to 5 seconds each time an individual pulse is detected in order to allow a longer count time at the location of the detected pulse, until it is determined whether the response indicates random background noise or detected contamination.

Several important factors affecting scan survey detection sensitivity are: instrument detection efficiency, background, size of the effective probe area, and the speed at which scan surveys are performed. For a given instrument, scan speed can be a critical factor as counting time is inversely proportional to scan speed. For instruments with larger detector faces, the scan speed is faster for a given rate of meter movement because a point on the surveyed surface remains beneath the window longer. To ensure that low levels of contamination can be detected, it is necessary that a maximum scan speed be mandated and that this speed be implemented during field measurements. As noted above, a typical scan speed for instruments in current use is 2 in./s (5 cm/s). However, the scan speed for a specific application should consider the instrument, probe, guideline value, and confidence level desired. The MARSSIM (NRC 1996) contains

guidance for determination of scan rates. It also suggests that an empirical method be used to verify scan rates. The equipment and method used in this determination may be incorporated into training for survey personnel to enhance their survey skills.

#### **4.2.5 Release Criteria**

The release of material from radiological areas shall be performed according to 10 CFR 835.1101. In these areas, material and equipment should be treated as radioactive material and should not be released from radiological areas to controlled areas if either of the following conditions exist:

- Measurements of accessible surfaces show that either the total or removable contamination levels exceed the values specified in Table 4.1
- Prior use suggests that the contamination levels on the inaccessible surfaces are likely to exceed the values specified in Table 4.1.

Wire rope and electronic gear with cooling fans are examples of equipment that are difficult to survey and require special procedures to be released from contaminated and airborne radioactivity areas. Additional release criteria can be found in Section 4 of the DOE Radiological Control Manual (DOE, 1994I).

It may be noted that Appendix D of 10 CFR 835 allows that surface radioactivity values be averaged over 1 m<sup>2</sup> provided that the activity in any 100 cm<sup>2</sup> is not more than three times the specified value.

The material release methodology has four main components: material evaluation, scan survey for fixed contamination, large-area wipe survey for removable contamination (described above), and statistical survey for fixed contamination. The material process involves consideration of the previous known uses of the material, as well as typical uses and the environment in which the material was used. Material evaluation places the material into one of two categories: not potentially contaminated or potentially contaminated.

Nonradioactive material can be released without an instrument survey if its documented history ensures

- That it has never been used or stored in an area controlled for contamination purposes (i.e., a Contamination Area, High Contamination Area, or Airborne Radioactivity Area)
- That it has never come into contact with unsealed radioactive material
- That it has not been stored or used in a Radiological Buffer Area (RBA) surrounding a Contamination Area, High Contamination Area, or Airborne Radioactivity Area.

This material may be considered to be not contaminated and an instrument survey is not necessary according to the DOE Radiological Control Manual (DOE, 1994I). A material history release form should be used to document the release of material that is known to be free of contamination by its history of use. If the material history release form cannot be completed, or if the history of the material is unknown, an instrument survey must be made of the material. Material released from RBAs around Contamination Areas, High Contamination Areas, or Airborne Radioactivity Areas should also be evaluated using an instrument survey.

The material evaluation process should also consider the nuclides to which the material was potentially exposed. If the material was exposed to significant quantities of nuclides that are difficult to detect, including tritium,  $^{14}\text{C}$ ,  $^{125}\text{I}$ , or  $^{129}\text{I}$ , an appropriate survey methodology should be applied.

#### **4.2.6 Plutonium Contamination Detection**

The detection and measurement of plutonium contamination is necessary to ensure control of contamination and compliance with DOE requirements. Typically, detection of plutonium contamination has been performed using survey instruments that detect the alpha activity. Routinely used health physics instruments (i.e., alpha survey instruments) may not be adequate for some D&D operations. Self-absorption of plutonium alpha particles within the source or in an irregular surface area may require the use of special

X-ray and low-energy photon instruments (e.g., a NaI detector). The NaI detector should also be used to detect plutonium contamination that has been painted over.

Discussions of methods used to detect plutonium contamination for past D&D operations can be found in publications by Umbarger (1982) and West et al. (1991). Umbarger reported on nondestructive assay techniques (including portable field instrumentation and laboratory-based methods) for sorting waste in low-level (class A) and TRU waste. Portable field instruments included the field FIDLER (i.e., thin NaI detector), phoswich detector (i.e., thin NaI detector coupled with a thicker CsI detector), ZnS alpha scintillation detector, a portable multichannel analyzer, and a hand-held gamma-ray spectrometer gun. The advantage of a phoswich detector over a NaI detector is its lower operating background. In the field, the phoswich detector has a detection limit of  $<1$  nCi/g. The ZnS scintillation detector had a detection limit of 25 pCi/g for gross alpha counting. Laboratory-based systems include active and passive gamma-ray spectroscopy, passive neutron detection, and pulsed portable neutron generator interrogation.

During the decommissioning of a mixed-oxide fuel fabrication facility, West et al. (1991) used a nondestructive assay system to provide criticality safety monitoring, track the plutonium inventory, provide measurement of decontamination effectiveness, and provide quantitative characterization/assay of the waste. The system consisted of an integrated set of two passive neutron networks, two pulsed active neutron units, a high-resolution gamma spectrometer [high-purity germanium (HPGe)], and a neutron-coincidence counting unit. Waste determined to be less than 10 nCi/g was certified as class A low-level waste (LLW).

#### **4.2.7 ALARA Guidelines**

Contamination levels should be maintained ALARA to minimize the potential for the spread of contamination and to reduce the protective measures and equipment required. Control of radioactive material at the source and prevention of the generation of contamination are more effective and less costly than remediation. As an example, studies have shown that workers required to wear respiratory protection are almost 25% less efficient than those doing the same work without wearing respirators.

### **4.3 PERSONNEL CONTAMINATION CONTROL**

As described earlier, the purpose of contamination control is to prevent the ingestion or inhalation of plutonium by workers. This is primarily achieved by the engineered barriers discussed previously, containment, confinement, and ventilation control. Only if the primary controls fail or if there is a potential for personnel contamination during an activity are administrative controls such as protective clothing and respirators advisable.

#### **4.3.1 Monitoring Philosophy**

Monitoring the worker is necessary, not only to ensure that a potential intake is detected promptly and that the resulting internal dose is assessed, but to confirm the integrity of the engineered containment system and ensure the effectiveness of the overall radiation protection program.

There are several types of worker monitoring, some during and immediately following work with radioactive material and some scheduled for a later time at a preset frequency. This section addresses only methods of monitoring the worker at the workstation. Other methods are discussed in the section that deals with internal and external exposure controls.

Techniques to monitor the individual worker at the work site include:

- Frequent/routine surveys of gloves
- exit surveys
- nasal swipes
- personal air sampling.

#### **4.3.2 Monitoring Program**

Instrumentation shall be provided and persons entering a plutonium work station shall be required to survey themselves at established frequencies. The requirements for radioactive contamination control and monitoring are found in 10 CFR 835.404. As a minimum, workers should survey their gloves and coverall sleeves each time they are

withdrawn from a glove box (or similar containment system) and after each glove replacement or bag-out operation.

Personnel monitoring for contamination should be mandatory at the egress from controlled areas and should be conducted in a verifiable manner. Assurance should be provided that personnel are monitored prior to breaks, meals, or exits from the plant site. Portal monitors, hand-and-shoe counters, and/or portable survey instruments may be used for this purpose. If employees are instructed to perform self-monitoring, the equipment should be set up in a “go/no-go” mode and employees should be clearly instructed in the required actions to take if predetermined action levels are exceeded. Frequent audits should be performed to verify that controls are adequate. Limiting the number of egress points and controlling personnel movement can minimize the numbers of locations where positive control of personnel monitoring must be maintained.

#### **4.3.3 Protective Clothing**

Various types of protective clothing, including laboratory coats, shoe covers, gloves, coveralls, plastic or rubber suits, and air-purifying or atmosphere-supplying respiratory protective equipment, may be required for operations with transuranic radionuclides. The use of company-issue shoes and clothing for employees with work assignments in process areas can be a major aid in contamination control. Recently, some facilities are using disposable anti-contamination clothing. This may be a cost savings from a handling standpoint. However, disposal costs must be considered. Additionally, consideration should be given to the potential for heat stress.

##### **4.3.3.1 Requirements for Routine Operations**

As a minimum, personnel who handle or work with unsealed sources of plutonium should wear coveralls, gloves, and shoe covers. For inspections or visits, lab coats and shoe covers may be permissible in those same areas. When contaminated wet areas are to be entered, water-repellent (plastic or rubber) clothing should be worn. No personal outer clothing should be permitted under coveralls.

When working with unsealed plutonium sources or in glove boxes, hands should be protected by a minimum of two barriers, for example, at least one pair of surgeon's gloves and one pair of glove-box gloves. Where manual dexterity is not required and the work involves a potential for piercing one or both layers of rubber gloves, leather gloves

should be worn over the surgeon's gloves. Automated methods should be considered for replacing routine manual methods that have a high risk of piercing the gloves.

Protective clothing should be removed at the exit to radiologically controlled areas and personnel monitoring for contamination performed. If for some reason this is not practical, the movement of personnel should be strictly controlled from the exit area to a location where protective clothing can be removed.

#### **4.3.3.2 Requirements for Special Maintenance**

For special maintenance work that involves significant quantities of plutonium, a double barrier concept should be implemented. An example of minimum requirements for protective clothing is provided below:

- Two pairs of coveralls (and sometimes a plastic suit)
- canvas boots taped to the inner pair of coveralls, with rubber boots over the canvas boots
- one pair of surgical gloves taped to the inner coveralls, with a leather, cotton, or rubber outer pair of gloves
- respiratory protective device with hood taped to respirator.

To create a double barrier between the source and all extremities, surgeon gloves should be worn in addition to the glove-box gloves.

In general, black Neoprene gloves are the standard glove-box glove and the most economical to use where process conditions do not produce rapid glove deterioration. However, alpha particles from surface dust layers can induce surface cracking in black Neoprene. Hypalon 0 is more resistant to surface cracking, acid deterioration, and ozone effects, and this characteristic will, in many cases, make Hypalon gloves the most economical, despite their higher unit cost.

In recent years many new types of glove-box gloves have been developed. Glove usage should be tailored to the particular needs of the job. For processes that require maximum dexterity, the 0.014-in. (0.038-cm) Neoprene gloves are still superior. Coated Hypalon gloves are superior to Neoprene for glove-box process operations that involve nitric acid or ozone levels that may cause deterioration. Ethylenepropylenediamine monomer (EPDM) gloves are used in some facilities and have good flexibility and are resistant to degradation caused by radiation and ozone. Greenhalgh et al. (1979) reported that

Hypalon and EPDM gloves have greater than 30 times the longevity of Neoprene in low-level ozone concentration atmospheres. Viton gloves have proven to have a longer life than Neoprene gloves under many operating conditions, but suffer somewhat from stiffness. Where high gamma radiation levels are encountered, lead-loaded gloves may be necessary. However, their stiffness and workers' loss of manual dexterity should be considered in determining their influence on work efficiency and the total dose received.

Persons who perform operations that involve microspheres of  $^{238}\text{Pu}$ , coated or uncoated, should be aware that the heat generation of a single 100- $\mu\text{m}$ - to 200- $\mu\text{m}$ -diameter sphere can melt through glove material. In addition, containment of a quantity of microspheres, especially coated microspheres, is difficult because of electrostatic repulsion.

Microspheres have been observed climbing the walls of a glass beaker and spreading throughout a glove box.

Glove storage problems occur occasionally. Experiments and static tests have not provided an adequate explanation of the sporadic problems that have been encountered. Test results in which gloves were stored under different lighting conditions (ultraviolet and fluorescent) and under stressed conditions (creased or bent) have not been consistent. Tests of gloves seem to indicate that glove degradation is caused by the combined effect of ionizing radiation, ozone, and lighting. The glove inventory should be rotated to prevent the inventory from becoming outdated while on the shelf.

All gloves in normal use at plutonium processing installations should be inspected prior to each use. All operating personnel should perform contamination self-surveys after every glove usage. The glove inspections should be made each time by the same team of trained individuals, and the condition of each glove should be recorded so that glove failures can be anticipated and preventative measures can be taken. The development of a statistical basis for establishing the frequency of glove changes should be considered because such a basis may be cost-effective. For example, the change-out frequency could be planned so that gloves are changed at some fraction of the mean time between failures or more preferably some fraction of the minimum time between failures. This type of change-out program could also minimize personnel doses and potential contamination spread incidents associated with too-frequent glove replacement. This procedure may require that each glove use be categorized. A routine replacement program will not replace an inspection program, but it is a supplement to the inspections. The inspector's surgeon gloves should, of course, be surveyed after the inspection of each glove-box glove. Gloves that are in questionable condition should be changed without delay.



Gloves that are not in use for the remainder of that shift should be capped off with a glove cover or plastic bag. Gloves not in use should be stored inside the glove box in such a manner that they do not interfere with operations.

#### **4.3.4 Respiratory Protection**

Respiratory protection should be readily available. Respiratory protective equipment should be used for all bag-out operations, bag and glove changes, and any situation involving a potential or actual breach of confinement. Alternatively, the operation could be performed in a glovebag to maintain confinement. In any case, protection, in the form of air-purifying or atmosphere-supplying respirators, should be considered whenever concentrations of radionuclides in the air are likely to exceed 10% of the Derived Air Concentration (DAC). For good performance, the respirator must fit closely on the facial contours and make an impenetrable seal so that all air enters through the filter or is supplied by the breathing-air supply. ANSI Z88.2-1992 (ANSI, 1980b) describes qualitative and quantitative tests that should be used to ensure that the respirator fits the individual; only the quantitative test should be used for verification of respirator fit at plutonium facilities. Respirator fit tests should be performed annually.

The respiratory protective device selected should provide a protection factor appropriate for the air concentration anticipated. ANSI Z88.2 provides protection factors guidance.

Air-supplied hoods are becoming more popular because a fitting is not required and facial hair does not prohibit their use. Protection factors greater than 1000 have been determined with air-supplied hoods. All respirators, including air-supplied hoods, require approval. While NIOSH approves most respirators, some respirator types in use at DOE facilities are not part of the NIOSH testing program. These are tested and approved by DOE's Los Alamos National Laboratory.

#### **4.3.5 ALARA Guidelines**

The total dose to an individual and the collective dose to the work force should be ALARA. When applied to personnel contamination or internal intakes, this generally means less-than-detectable dose with the best available commercial technology.

#### **4.3.6 Release Criteria**

The decision to release personnel with detectable plutonium contamination is made on a case-by-case basis. If the individual is injured and needs prompt medical attention, medical treatment will always take precedence, with compensatory measures made for the protection of medical personnel and facilities. If injuries are absent or do not require immediate attention, decontamination is preferable to ensure that the dose to the contaminated individual and the potential for inhalation by the victim and medical staff are minimized and the spread of contamination is prevented.

In a case where decontamination is incomplete due to injury to the skin or other reasons, the individual may be provisionally released with measures to prevent the spread of contamination.

### **4.4 Personnel Decontamination**

Skin decontamination should be performed by health physics technicians or other members of the health physics staff. The treatment and decontamination of wounds should be performed by medical staff.

Nonabrasive methods should be used for skin decontamination to protect the tissues from deeper contamination. Masking tape should be used to remove dry contamination. Wet decontamination should be used to remove residual contamination. The skin should be gently scrubbed with soap

and water. Household bleach may be applied as needed to decontaminate more effectively. The following procedure is recommended:

1. Survey the worker to determine the contaminated areas of the skin. Have the medical staff treat and decontaminate breaks in the skin.
2. Wipe loose contamination with a gauze sponge or cotton applicators dipped in mild anti-septic detergent. Do not spread contamination to uncontaminated areas.
3. Rub the skin with the applicators to produce good sudsing.
4. Use soft bristle scrub brushes for fingernails and other difficult-to-clean areas as long as the skin barrier is maintained intact. It may be difficult to decontaminate the cuticles and under the nails.
5. Dry the skin area with cleansing tissue.
6. After the skin is thoroughly dry, survey it for any remaining contamination.
7. If no contamination is detected, apply a good-quality hand cream to prevent chapping.

Another effective nonabrasive decontamination method involves placing the contaminated hand in a cotton glove and then a Latex glove (causing the hand to perspire).

The decontamination factor is the ratio of the initial contamination level to the contamination level after decontamination methods are applied, as determined by survey instrument readings.

Nonabrasive methods should be repeated until the decontamination factor between washes drops below 2 or 3 with significant contamination still remaining.

If contamination persists on the skin, a more abrasive decontamination method may be necessary. The decision to proceed with a more abrasive method should be based on the effectiveness of the decontamination. An abrasive soap should be applied with a moist gauze sponge or soft handbrush while rubbing the skin to develop a soapy lather. Care should be exercised to prevent damage to the skin surface. If contamination persists after using the abrasive soap, potassium permanganate ( $\text{KMnO}_4$ ) and sodium acid sulfite should be considered. Page 196 of the 1970 edition of the Radiological Health Handbook gives details on the use of  $\text{KMnO}_4$  and sodium acid sulfite.

Liberal irrigation with lukewarm water or saline solution is recommended for eye, nose, and mouth contamination. These procedures are performed by the medical staff to remove contamination.

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## **5.0 INTERNAL DOSIMETRY**

Internal dosimetry is an essential part of a quality health physics program at every facility where plutonium is handled or processed. The purpose of an internal dosimetry program is to monitor workplace activities, to assess accidental or inadvertent intakes of radioactive material, and to conduct internal dose assessments from bioassay measurement data.

It is DOE policy that facilities are designed, operated, and remediated to prevent intakes of radioactive materials. Radiological controls for the workplace should ensure that radionuclides are contained and handled properly, and that intakes, if they occur at all, are negligible to the extent achievable with state-of-the-art technology. In spite of excellent design and operation policies, inadvertent intakes of radioactive material can occur as a result of equipment malfunction, failure to follow procedures, or the unanticipated presence of radioactive material.

Experience has shown that the most common route for inadvertent plutonium intake is inhalation. Intakes can also occur by accidental ingestion or by wound contamination. Surveillance programs should be designed to rapidly detect a release in the event of a loss of radioactive material containment. Internal dosimetry programs should be tailored to the needs of each plutonium-handling facility so that inadvertent intakes are discovered and quantified and workers' dose equivalents are determined by appropriate methods.

When workers are inadvertently exposed to radioactive material, appropriate corrective action should be taken to ensure that control and containment have been re-established. Prompt detection by routine workplace monitoring practices is essential to regaining control after any contamination spread or loss of containment. Prompt workplace indications of potential intake are also crucial to start special bioassay monitoring for intake and dose assessment. An early assessment of the probable severity of an intake and its corresponding dose, preferably within the first two hours of the intake, is needed for decisions on dose reduction therapy and event reporting. For plutonium and americium intakes, the bioassay data necessary for final dose assessment may require long periods of time (many months) to obtain. Until such data become available, ongoing preliminary assessments of intake and dose may be necessary to provide guidance for the administrative and medical management of the workers.

## **5.1 INTERNAL DOSE EVALUATION PROGRAM**

Internal doses are not directly measured but are estimated or calculated based on knowledge of the material to which a worker may be exposed and its known or assumed biokinetic behavior. The common approach to internal dosimetry is to calculate an occupational intake based on worker bioassay measurements or workplace air-sample data and assumed breathing rates. Once an intake is calculated, appropriate internal dose equivalents to organs and tissues of concern can be estimated by using fundamental dosimetry principles, by various intake-to-dose conversion factors, which incorporate assumed biokinetic models, or by an appropriate computer code. Intake-to-dose conversion factors can be found in Federal Guidance Report No. 11 (EPA, 1988b) or ICRP Publication 30 (ICRP, 1979, 1988b). Further discussion on intake and dose assessment is provided in Section 5.8.

Participation in internal dose evaluation programs is required by DOE for conditions identified in 10 CFR 835.402(c) (DOE, 1993c). The internal dose evaluation program must address both general workplace conditions and individual intakes.

Workplace conditions are monitored through air sampling programs as well as contamination surveys. For work that can have variable or changing conditions, more intensive surveillance may be required, using supplemental portable air samplers, continuous air monitors, or personal air samplers.

Individual worker monitoring for intakes is commonly performed using bioassay procedures. Bioassay monitoring includes both direct (in vivo) measurements of radioactivity in the body and indirect (in vitro) measurements of material excreted or removed from the body. Refer to section 5.7.4 for information on assessing internal exposure from air supply data.

10 CFR 835.402(c) (DOE, 1993c) specifies the requirements for participation in a radiological bioassay program. Because most plutonium facilities have a high degree of radiological control and containment for plutonium, chronic exposure to levels of occupational concern is unlikely and it is not considered likely that a worker would incur more than one unplanned intake in a year. Thus, participation in a bioassay program is generally based on the possibility that a single

intake causing a dose in excess of 100-mrem committed effective dose equivalent (CEDE) might occur. Bioassay is also required if an intake is suspected for any reason.

Indications of intake include (but are not limited to) detection of facial or nasal contamination, air monitoring or sampling that indicates internal exposure, or any wound in which contamination is detected or suspected (See Section 5-9 for internal dosimetry recommended indicator and action levels.) The most common internal exposure monitoring program for workers is the bioassay monitoring program, which must be designed for the specific nuclides and forms of material at a particular facility. Likely candidates for internal exposure monitoring include personnel who may be routinely exposed to surface or airborne contamination, or those identified by the foregoing workplace indicators.

Workplace monitoring for potential internal exposures is performed to verify the adequacy of containment and work practices. This monitoring includes air sampling, continuous air monitoring, personal contamination surveys, and workplace contamination surveys. Facilities are to be designed and operated to minimize internal exposure. Details regarding workplace monitoring and control practices are discussed in Section 4.0.

### **5.1.1 Performance Capabilities for Internal Exposure Monitoring**

Bioassay monitoring programs must be capable of showing compliance with the 5-rem/year stochastic and 50-rem/year nonstochastic dose limits of 10 CFR 835.202 (DOE, 1993c). 10 CFR 835.402(c)(1) (DOE, 1993c) identifies 100-mrem CEDE and/or 5 rem CDE to any organ or tissue for all likely intakes as a level above which workers must participate in a bioassay program. Therefore, ideally, such bioassay monitoring programs should be capable of detecting those levels. In fact, this is not technically achievable for most routine plutonium bioassay programs. In order to meet this requirement, reliance must be placed on workplace monitoring to identify potential intakes at the time they occur so that special bioassay monitoring can be initiated. Routine, periodic bioassay measurements have little chance of detecting a CEDE of 0.1 rem and can even have difficulty showing compliance with dose limits.



Performance capabilities for bioassay and internal dosimetry programs can be expressed as the minimum detectable dose, based on some combination of minimum detectable activity and frequency of measurement or time post-intake at which the measurement is made. The term "minimum detectable dose" is preferred over any variants of the occasionally encountered terms "dose-missed" or "potentially undetected dose," which were usually defined as the same thing. The connotation of the latter terms is that of an actual intake which was not detected, whereas the intent was to define a measure of program sensitivity to doses that might have gone undetected had an intake occurred. The preferred term "minimum detectable dose" (MDD) ties the concept to the recognized terminology of minimum detectable activity (MDA).

The MDD for a bioassay monitoring program must meet the aforementioned dose limit requirements of 10 CFR 835.202. A design goal of 100-mrem CEDE from all intakes of similar nuclides in a year is desirable but unrealistic for a routine program. To meet these requirements, bioassay programs should have measurement sensitivities (i.e., MDAs for bioassay measurements) established based on the material to which workers might be exposed. Examples of such sensitivities are given in Tables 5.1 and 5.2 for pure  $^{239}\text{Pu}$  monitored by urinalysis and fecal analysis, respectively. Table 5.3 provides an example of the  $^{241}\text{Am}$  sensitivity required for monitoring a mixture of weapons-grade plutonium, aged 5 years for ingrowth at time of intake. These tables illustrate the difficulty in relying on routine bioassay to demonstrate compliance with the limits and design goal.

The problem is simply that the measurement technology is not available to provide the sensitivities required for the 100-mrem goal using routine, periodic measurements at reasonable frequencies. Therefore, because the goal of 100-mrem CEDE cannot be met through routine bioassay, the radiation protection organization should take the following administrative actions:

- ensure that adequate control measures are applied to prevent intakes
- document the adequate control measures for auditing purposes

- upgrade bioassay measurement systems and workplace monitoring practices to provide state-of-the-art measurements
- ensure that internal dose assessments use state-of-the-art technology.
- ensure workplace monitoring programs are designed to identify potential intakes.

**Table 5.1.** Urine Bioassay Goals<sup>(a)</sup> for <sup>239</sup>Pu

Days Post-Intake	Class W Inhalation			Class Y Inhalation		
	Urine	Dose	100-mrem	Urine	Dose	100-mrem
	Intake	Limit	CEDE	Intake	Limit	CEDE
	Retention Fraction	Goal, <sup>(b)</sup>	Goal, <sup>(c)</sup>	Retention Fraction	Goal, <sup>(b)</sup>	Goal, <sup>(c)</sup>
	(IRF)	dpm	dpm	(IRF)	dpm	dpm
1	2.36E-04	3.35E+00	1.22E-01	1.28E-05	4.67E-01	9.23E-03
2	1.61E-04	2.29E+00	8.33E-02	8.71E-06	3.18E-01	6.28E-03
7	3.29E-05	4.68E-01	1.70E-02	1.69E-06	6.17E-02	1.22E-03
14	2.18E-05	3.10E-01	1.13E-02	1.09E-06	3.98E-02	7.86E-04
30	1.73E-05	2.46E-01	8.95E-03	8.60E-07	3.14E-02	6.20E-04
60	1.34E-05	1.90E-01	6.93E-03	7.15E-07	2.61E-02	5.15E-04
90	1.15E-05	1.63E-01	5.95E-03	6.89E-07	2.52E-02	4.97E-04
180	8.31E-06	1.18E-01	4.30E-03	7.19E-07	2.63E-02	5.18E-04
365	4.84E-06	6.88E-02	2.50E-03	7.97E-07	2.91E-02	5.74E-04
730	2.45E-06	3.48E-02	1.27E-03	8.85E-07	3.23E-02	6.38E-04
1825	1.63E-06	2.32E-02	8.43E-04	8.82E-07	3.22E-02	6.36E-04
3650	1.54E-06	2.19E-02	7.97E-04	7.38E-07	2.69E-02	5.32E-04
7300	1.38E-06	1.96E-02	7.14E-04	6.09E-07	2.22E-02	4.39E-04
18250	1.00E-06	1.42E-02	5.17E-04	4.38E-07	1.60E-02	3.16E-04

(a) The goals reflect the activity in a 24 hour urine void corresponding to either a 50 rem CDE or a 0.1 rem CEDE.

(b) Calculated as Goal (dpm) = Intake \* IRF \* 2220 dpm/nCi, where Intake (nCi) is the 50 rem CDE dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from Federal Guidance Report No. 11 (EPA, 1988b), Table 2.1 is shown below:

	Class W,	Class Y,
	<u>rem/nCi</u>	<u>rem/nCi</u>
Bone Surface	7.81	3.04

(c) Calculated as Goal (dpm) = Intake \* IRF \* 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem CEDE dose threshold/dose conversion factor and IRF is the intake retention fraction.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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The dose conversion factor (committed dose per unit intake) derived from Federal Guidance Report No. 11 (EPA, 1988b), Table 2.1 is shown below:

	Class W, Class Y,	
	<u>rem/nCi</u>	<u>rem/nCi</u>
DCF	0.429	0.308

**Table 5.2.** Fecal Bioassay Goals<sup>(a)</sup> for <sup>239</sup>Pu

Days Post- Intake	Class W Inhalation			Class Y Inhalation		
	Fecal	Dose	100-mrem	Fecal	Dose	100-mrem
	Intake	Limit	CEDE	Intake	Limit	CEDE
	Retention Fraction <sup>(b)</sup> (IRF)	Goal, <sup>(c)</sup> dpm	Goal, <sup>(d)</sup> dpm	Retention Factor <sup>(b)</sup> (IRF)	Goal, <sup>(c)</sup> dpm	Goal, <sup>(d)</sup> dpm
1	1.05E-01	1.49E+03	5.43E+01	1.27E-01	4.64E+03	9.15E+01
2	1.34E-01	1.90E+03	6.93E+01	1.56E-01	5.70E+03	1.12E+02
7	6.41E-03	9.11E+01	3.32E+00	5.52E-03	2.02E+02	3.98E+00
14	1.23E-03	1.75E+01	6.37E-01	1.73E-04	6.32E+00	1.25E-01
30	9.58E-04	1.36E+01	4.96E-01	1.34E-04	4.89E+00	9.66E-02
60	6.34E-04	9.01E+00	3.28E-01	1.29E-04	4.71E+00	9.30E-02
90	4.21E-04	5.98E+00	2.18E-01	1.23E-04	4.49E+00	8.87E-02
180	1.26E-04	1.79E+00	6.52E-02	1.09E-04	3.98E+00	7.86E-02
365	1.39E-05	1.98E-01	7.19E-03	8.46E-05	3.09E+00	6.10E-02
730	2.50E-06	3.55E-02	1.29E-03	5.14E-05	1.88E+00	3.70E-02
1825	1.63E-06	2.32E-02	8.43E-04	1.19E-05	4.35E-01	8.58E-03
3650	1.54E-06	2.19E-02	7.97E-04	1.36E-06	4.97E-02	9.80E-04
7300	1.38E-06	1.96E-02	7.14E-04	6.15E-07	2.25E-02	4.43E-04
18250	1.00E-06	1.42E-02	5.17E-04	4.39E-07	1.60E-02	3.16E-04

- (a) The goals reflect the activity in a 24 hour fecal sample corresponding to either a 50 rem CDE or a 0.1 rem CEDE.
- (b) IRF values obtained from GENMOD Computer Code (Version 3.0.3).
- (c) Calculated as Goal (dpm) = Intake \* IRF \* 2220 dpm/nCi, where Intake (nCi) is the 50 rem CDE dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from Federal Guidance Report No. 11 (EPA, 1988b), Table 2.1 is shown below:

Class W, Class Y,

	<u>rem/nCi</u>	<u>rem/nCi</u>
Bone Surface	7.81	3.04

- (d) Calculated as Goal (dpm) = Intake \* IRF \* 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem CEDE dose threshold/dose conversion factor and IRF is the intake retention fraction.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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The dose conversion factor (committed dose per unit intake) derived from Federal Guidance Report No. 11 (EPA, 1988b), Table 2.1 is shown below:

	Class W, Class Y,	
	<u>rem/nCi</u>	<u>rem/nCi</u>
DCF	0.429	0.308

**Table 5.3.** In Vivo Lung Measurement Bioassay Goals<sup>(a)</sup> for <sup>241</sup>Am as an Indicator of Aged Weapons-Grade Plutonium<sup>(b)</sup>

Days Post-Intake	Class W Inhalation		Class Y Inhalation	
	Dose Limit	100-mrem	Dose Limit	100-mrem
	Goal, <sup>(c)</sup>	CEDE Goal, <sup>(d)</sup>	Goal, <sup>(c)</sup>	CEDE Goal, <sup>(f)</sup>
	nCi <sup>241</sup> Am	nCi <sup>241</sup> Am	nCi <sup>241</sup> Am	nCi <sup>241</sup> Am
1	7.2E-02	2.9E-03	1.8E-01	4.1E-03
2	6.1E-02	2.5E-03	1.6E-01	3.5E-03
7	4.7E-02	1.9E-03	1.3E-01	2.9E-03
14	4.3E-02	1.7E-03	1.3E-01	2.9E-03
30	3.5E-02	1.4E-03	1.3E-01	2.8E-03
60	2.4E-02	9.7E-04	1.2E-01	2.7E-03
90	1.6E-02	6.6E-04	1.2E-01	2.6E-03
180	5.1E-03	2.1E-04	1.1E-01	2.4E-03
365	4.6E-04	1.9E-05	8.9E-02	2.0E-03
730	3.8E-06	1.5E-07	6.3E-02	1.4E-03
1825	1.3E-11	5.1E-13	2.5E-02	5.6E-03
3650	6.9E-13	2.7E-14	8.2E-03	1.8E-04
7300	4.6E-14	1.8E-15	3.5E-03	7.8E-05
18250	5.0E-18	2.0E-19	3.0E-03	6.7E-05

(a) Calculated using the CINDY Code (Version 1.3) (Streng et al., 1992).

(b) Defined as a Pu mixture consisting of, by weight %, 93% <sup>239</sup>Pu, 6.1% <sup>240</sup>Pu, 0.8% <sup>241</sup>Pu, 0.05% <sup>238</sup>Pu, and 0.05% <sup>242</sup>Pu, with 5 years allowed for <sup>241</sup>Am ingrowth.

(c) Intake of 4.7-nCi Pu-α, 38-nCi <sup>241</sup>Pu, and 0.34-nCi <sup>241</sup>Am, giving a committed dose equivalent of 50 rem to the bone surfaces.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- (d) Intake of 0.19-nCi Pu- $\alpha$ , 1.5-nCi  $^{241}\text{Pu}$ , and 0.014-nCi  $^{241}\text{Am}$ , giving a committed effective dose equivalent of 0.1 rem.
- (e) Intake of 12-nCi Pu- $\alpha$ , 97-nCi  $^{241}\text{Pu}$ , and 0.86-nCi  $^{241}\text{Am}$ , giving a committed dose equivalent of 50 rem to the bone surfaces.
- (f) Intake of 0.27-nCi Pu- $\alpha$ , 2.2-nCi  $^{241}\text{Pu}$ , and 0.019-nCi  $^{241}\text{Am}$ , giving a committed effective dose equivalent of 0.1 rem.

All confirmed occupational intakes of plutonium, regardless of magnitude, should be assessed. The results of all bioassay and other measurements needed to support the quality of measurements and dose assessment should be recorded and maintained. The recording and reporting requirements for internal dosimetry data are set forth in Section 3.7 of this report; however, the following is a summary list of internal dosimetry information for which recording is required:

- Total CEDE from all intakes during a year
- committed dose equivalent (CDE) to organs or tissues of concern from all intakes during a year
- magnitude of intake for each radionuclide during a year
- data necessary to allow subsequent verification, correction, or recalculation of doses
- gestation period dose equivalent to the embryo/fetus from intake by the declared pregnant worker during the entire gestation period.



Radiation exposure records programs must also provide for the summation of internal and external doses, as required by 10 CFR 835.702 (DOE, 1993c). While the summation process is not necessarily performed under a site internal dosimetry program, it behooves the program to recognize what is required. The following summations are identified by 10 CFR 835.702(c)5):

- Total effective dose equivalent (TEDE) defined as the summation of effective dose equivalent (deep dose equivalent) from external exposure and the CEDE
- summation of the effective dose equivalent (deep dose equivalent) from external exposure and the CDE to organs or tissues of concern
- cumulative TEDE received from external and internal sources while employed at the site or facility, since January 1, 1989
- for the embryo/fetus of a declared pregnant worker, the summation of the deep dose equivalent to the mother from external exposure during the entire gestation period and the gestation period dose equivalent to the embryo/fetus from intakes by the mother during the entire gestation period.

Doses should be calculated and recorded for any confirmed plutonium intake. What constitutes a confirmed intake is discussed in Section 5.7. Along with the doses, supporting records must be maintained, including the bioassay data, assumptions, biokinetic models, and calculational methods used to estimate the doses. These may be included in letter-report dose assessments, databases, technical basis documents, and similar records, either singly or in combination.

#### **5.1.2 Protection of the Embryo/Fetus, Minors, and Members of the Public**

The TEDE limit for the embryo/fetus of a declared pregnant worker is 0.5 rem for the entire gestation period, defined as the summation of external dose received and internal dose received during the gestation period (not the 50-year committed internal dose). Internal exposure monitoring is required if an intake is likely to result in more than 10% of that limit (i.e., 50 mrem for the gestation period). As discussed in more detail in Section 5.6., providing adequate protection to keep the mother's intakes below the occupational limits will also provide adequate protection for the embryo/fetus. Thus, special bioassay for plutonium or americium related to pregnancy is not required. As a matter of caution, some sites try to obtain baseline bioassays as soon as a pregnancy is declared, with another baseline bioassay following the end of pregnancy. Some sites also offer to restrict pregnant workers from jobs with relatively high potential for occupational intakes.

Minors and members of the public are limited by 10 CFR 835.207 and 10 CFR 835.208 (DOE, 1993c) to a TEDE of 0.1 rem/year. Internal exposure monitoring is required if an intake is likely to result in 50% of that limit (0.05 rem). As noted in Section 5.1.1, because bioassay monitoring is not likely to be sufficiently sensitive to identify such intakes on a routine basis, enhanced workplace surveillance or restriction of access may be required.

## **5.2 CHARACTERIZATION OF INTERNAL HAZARDS**

Plutonium can be encountered in a wide range of mixtures, e.g., a pure isotope in a standard solution, a highly variable combination of isotopes in so-called "weapons grade" or "fuels grade" Pu, or commercial spent fuel. In addition, the age of a mixture significantly affects its isotopic composition. As a typical weapons or fuels grade mixture ages, the  $^{241}\text{Pu}$  decays to  $^{241}\text{Am}$ . Although the mass changes may be quite small, the overall result can be a significant build-up of  $^{241}\text{Am}$  radioactivity with time. This buildup can make the mixture somewhat easier to detect by in vivo methods. Table 5.4 shows some example plutonium mixtures which might be encountered in DOE facilities. Isotopically pure forms of radionuclides can also be encountered. Table 5.5 demonstrates the impact of aging on the activity composition of two mixtures. The

composition of plutonium in the facility can significantly affect the design and capabilities of an internal dosimetry program. As part of the program technical basis, the plutonium mixtures need to be determined. In addition, determinations should be made at the time of identified incidents of potential intake. Methods for such determination may include radiochemical analysis or chemistry followed by mass spectrometry.

The physical-chemical form of plutonium also affects the internal hazard posed. Oxides of plutonium tend to exhibit inhalation class Y behavior, whereas other compounds such as nitrates are assigned class W by the ICRP. However, as noted in Section 2.4.1, extremes have been observed with regard to both highly soluble and highly insoluble forms, leading to the good practice of performing dissolution rate (i.e., solubility) tests on standard materials in a facility.

As plutonium ages in a residual, loose contamination form, such as might be found in old duct work, glove boxes, or other such components, it can be expected to undergo slow oxidation to a more insoluble form. Thus, class Y forms of plutonium may be reasonable assumptions of what to expect during many decommissioning operations.

Particle size is an important consideration for inhalation exposures. The normal practice for an aerosol is to identify the activity median aerodynamic diameter and its associated particle-size distribution. Particle sizes of 10  $\mu\text{m}$  or less are considered respirable. The common practice is to assume a 1- $\mu\text{m}$  particle size for dosimetry purposes because actual particle size information is usually lacking. Particle size data are most readily obtainable for chronic exposure situations. Unless representative air sampling is performed in the immediate proximity of a worker during abnormal working conditions, the practical likelihood of obtaining good particle-size information is slim.

**Table 5.4.** Example Plutonium Isotope Mixtures Immediately Post-Separation, wt%

Isotope	Weapons-Grade	Fuels-Grade	Spent Commercial
	Plutonium	Plutonium	Fuel
	(6% $^{240}\text{Pu}$ Mixture)	(12% $^{240}\text{Pu}$ Mixture)	(25% $^{240}\text{Pu}$ Mixture)
$^{238}\text{Pu}$	0.05	0.10	1.49
$^{239}\text{Pu}$	93.0	84.4	59.50
$^{240}\text{Pu}$	6.1	12.4	23.98
$^{241}\text{Pu}$	0.8	3.0	10.33
$^{242}\text{Pu}$	0.05	0.1	4.0
$^{241}\text{Am}$	0.0	0.0	0.0

**Table 5.5.** Activity Composition with Age for Reference 6% and 12%  $^{240}\text{Pu}$  Mixtures

Isotopic Component	Reference 6% Pu Mix <sup>(a)</sup>		Reference 12% Pu Mix <sup>(a)</sup>	
	Fresh	Aged	Fresh	Aged
<u>Specific Activity in Mixture, Ci/g</u>				
$^{238}\text{Pu}$	8.6E-3	8.2E-3	1.7E-2	1.6E-2
$^{239+240}\text{Pu}$	7.2E-2	7.2E-2	8.0E-2	8.0E-2
$^{241}\text{Pu}$	8.2E-1	8.2E-1	3.1E+0	2.4E+0
$^{242}\text{Pu}$	2.0E-6	2.0E-6	3.9E-6	3.9E-6
$^{241}\text{Am}$	5.3E-5	5.8E-3	2.0E-4	2.2E-2
Pu-alpha	8.1E-2	8.1E-2	9.7E-2	9.6E-2
Total alpha	8.1E-2	8.1E-2	9.7E-2	1.2E-2
<u>Activity Ratios</u>				
$^{239+240}\text{Pu} : ^{241}\text{Am}$	1.2E+3	1.2E+1	4.1E+2	3.7E+0
Pu-alpha : $^{241}\text{Am}$	1.5E+3	1.4E+1	4.9E+2	4.4E+0
$^{241}\text{Pu} : ^{239+240}\text{Pu}$	1.2E+1	9.1E+1	3.8E+1	3.0E+1

(a) % = nominal  $^{240}\text{Pu}$  weight percent in mixtures.

Fresh = 2 weeks of  $^{241}\text{Am}$  ingrowth following separation.

Aged = 5 years of  $^{241}\text{Am}$  ingrowth following separation.

### 5.3 SCOPE OF BIOASSAY PROGRAM

The relatively low annual limit on intake of plutonium renders its radiation hazard substantially more restrictive than its industrial hygiene or chemical toxicity hazard. Thus, internal radiation dose or intake monitoring is the appropriate focus of bioassay monitoring.

### 5.3.1 Classification of Bioassay Measurements

Bioassay measurements can be classified according to the primary reason for their performance. This is a useful practice for historically documenting why a worker participated in a bioassay program. Numerous reasons for bioassay measurements may be defined for specific facilities; some suggested common classifications are as follows:

- **Baseline measurements** are used to establish a pre-exposure condition, either for a new employee or as a result of a new work assignment. The Radiological Control Manual (DOE, 1994I) recommends baseline measurements if workers are considered likely to receive intakes resulting in greater than 100-mrem CEDE. It is a good practice to perform such measurements for newly hired employees, intra-company transferees, or workers transferred from facilities where bioassay measurements may not have been required. In addition, baseline measurements can verify workers' status for special work assignments. For plutonium bioassay, baseline measurements made before any occupational exposure can be expected to yield no detectable results using current technology.

Exempting workers from baseline bioassay implies accepting any detectable results as likely attributable to current occupational exposure. However, requiring baseline measurements can potentially impact the schedule of short-term jobs; the time required to obtain a chest count and a large-volume urine sample may add a day or two delay to entry procedures. Moreover, missing a baseline for a long-term employee who will be placed on a routine bioassay program is not likely to be as troublesome as not obtaining a baseline for a short-term worker who provides a termination sample that shows detection of plutonium after the worker has left the site and is difficult to reach for follow-up.

- **Routine, or periodic, measurements** are performed on a predetermined schedule (e.g., an annual or quarterly frequency).

- **Special bioassay measurements** are performed as follow-up to unusual routine results or suspected intakes (See Section 5.9 for recommended internal dosimetry indicator and action levels).
  
- **End of assignment or termination measurements** are performed following completion of specific work or at the time of termination of employment. The Radiological Control Manual (DOE, 1994I) recommends that workers who participate in bioassay programs have appropriate termination measurements.

Bioassay classification is important because the purpose of a sample may affect the collection and analysis or monitoring method chosen. For example, single-void urine samples are not adequate for routine monitoring of potential plutonium exposure, but can provide important information for dose-reduction therapy following a suspected intake; samples representative of excretion over a 24-hour period should be collected for quantitative intake and dose assessment. The date of sample collection (and possibly the time of collection) can be very important to special monitoring performed to assess intake. However, these are much less important with regard to periodic monitoring, for which measurements are not expected to show detectable activity and when any detection whatsoever is likely to initiate investigation and special bioassay.

### 5.3.2 Monitoring Requirements and Selection of Employees

Workers who are considered likely to have intakes resulting in excess of 100-mrem CEDE are required to participate in a bioassay program. However, because of the extensive radiological control practices for plutonium facilities, including a high degree of engineered barrier containment, no typical plutonium worker is likely to have intakes of 100-mrem CEDE or more. However, this should not be used as an excuse to exclude workers from routine bioassay. Although no one should be considered likely to have intakes resulting in 100-mrem CEDE, some workers are at significantly higher risk for incurring an intake than others and should be on routine bioassay.

The workers at highest risk of incurring an intake are the ones in closest contact with the material. Typically, these are the operators, maintenance, and health physics personnel handling plutonium or plutonium-contaminated objects in the course of routine glove-box, maintenance, or decommissioning operations. In the event of containment system failure, or failure respiratory protection devices, it is these workers who will most likely incur exposure and subsequent intake. These workers should be on a routine bioassay program designed to meet the requirements of 10 CFR 835 (DOE, 1993c) as a kind of safety net to identify intakes which might have gone undetected by workplace monitoring.

Other workers (e.g., supervisors, inspectors, observers, guards, and tour groups) who work in or visit a plutonium facility but are not directly working with the material or contaminated objects are at a substantially lower risk for incurring an intake. Although these people may not need to be on a routine bioassay program, they should be subject to participation in a special bioassay program if workplace indications suggest loss of control or containment.

### **5.3.3 Selection of Bioassay Monitoring Techniques**

Bioassay monitoring techniques fall into two broad categories, direct measurement of radioactive materials in the body (in vivo counting) and analysis of material removed from the body for laboratory in vitro analysis. In vivo counting includes measurements of the chest, lung, skeleton, liver, and wounds. In vitro measurements include urinalysis, fecal analysis, and occasionally analysis of tissue, sputum, or blood samples. Methods for in vitro analysis include liquid scintillation counting, fluorescence measurements, gamma spectrometry, chemical separation followed by electrodeposition, and counting with radiation detectors. Selby et al. (1994) provide a brief overview of bioassay techniques and capabilities. Further discussion of the techniques is provided below.

#### **5.3.3.1 In Vivo Counting**



Direct bioassay (in vivo counting) is the measurement of radiations emitted from radioactive material taken into and deposited in the body. Direct bioassay is appropriate for detection and measurement of photons emitted by plutonium and its decay products. Lung, wound, liver, and skeleton counting are examples of in vivo monitoring most commonly used for plutonium and its progeny. Whole body counting, commonly used for monitoring high-energy fission and activation products in the body, is ineffective for direct measurement of plutonium due to the very low energy of photons emitted from plutonium and its decay products unless the plutonium is intimately mixed in a high-energy photon-emitting matrix, such as spent fuel.

Some low-energy x-rays emitted by plutonium decay products are energetic enough to escape the body. When direct bioassay is used, the detection system should be calibrated for the radionuclides to be measured in the appropriate organs. All calibration procedures, calibration records, and quality control data should be maintained. Energies most commonly used for plutonium monitoring are the 17-keV L X-rays and the 60-keV gamma of  $^{241}\text{Am}$ . Mixtures of spent fuel material can lend themselves to whole body counting if the ratio of a readily detectable high-energy gamma-emitter (i.e.,  $^{137}\text{Cs}$ ) to plutonium is known.

A plutonium facility should have the capability to detect and assess depositions of plutonium in the lungs of radiation workers. The major objective of lung counting is to provide measurements of suspected intakes triggered by workplace monitoring results. Lung measurements should be made to provide an early estimate of the magnitude of the intake and resulting lung deposition.

Two methods have been used to detect plutonium in the lung: the L x-ray method and the americium-tracer method. The L x-ray method is based on the measurement of L X-rays following the decay of plutonium. This method provides a direct measurement of plutonium. The detection capability of the method may be on the order of tens of nanocuries for plutonium and requires an

accurate measurement of the chest wall thickness (because of the large attenuation of the low-energy X-rays by the rib cage and overlying tissues). Other problems that complicate the measurement of L X-rays are 1) the difference in attenuation in muscle and fat, 2) the possibility of nonuniform distribution of the plutonium in the lung, and 3) interferences from radionuclides in other organs or from other radionuclides in the lung.

The americium-tracer method has the advantage of better detection capability for some mixtures of plutonium. The typical MDA for  $^{241}\text{Am}$  lung counting is 0.1 to 0.2 nCi. The americium-tracer method depends on the plutonium/americium ratio, which must be independently determined or estimated for each intake. The detection level for this method with a plutonium/americium ratio of 15 is typically 2-nCi plutonium in the lung. The americium-tracer method also has the advantage of being less affected by attenuation in the chest wall or by variations in the muscle/fat ratio. However, it has the disadvantage of requiring an estimate of the plutonium/americium ratio, both initially and at long times post intake. This ratio may change over time because of ingrowth of  $^{241}\text{Am}$  as the decay product of  $^{241}\text{Pu}$  or because americium may naturally clear from the lungs and translocate among internal organs at a rate different than that for plutonium.

The most widely used systems for lung counting are high-purity germanium detectors, thin sodium-iodide detectors, phoswich detectors, and proportional counters. Multiple high-purity germanium detectors have advantages over the other detector systems because of their good resolution, allowing better identification of the radionuclide, better detectability, and better background prediction capability. The main disadvantages of germanium detector arrays are their higher cost relative to other types of in vivo detectors and their lower reliability. Germanium detectors also must be continuously cooled with liquid nitrogen.

Measurement equipment to detect and measure plutonium contamination in wounds should be available at all plutonium facilities. Instrumentation used for this purpose may include thin-crystal NaI(Tl), intrinsic germanium, or Si(Li) detectors. The detection level for plutonium wound measurements is typically 0.1 nCi for  $^{239}\text{Pu}$ . Correction for depth due to absorption of photons in the overlying tissues should be considered. Collimated detectors are useful for determining the location of the plutonium in wounds.

Estimates of the depth of plutonium contamination in a wound may be made using solid-state germanium or Si(Li) detectors to measure the relative absorption of the low-energy X-rays emitted by plutonium. Information about depth is important for determining whether tissue excision is necessary to remove the contamination.

#### **5.3.3.2 In Vitro Analysis**

The two most common forms of in vitro analysis are urinalysis and fecal analysis.

**Urinalysis.** Urine sampling provides useful information about the amount of plutonium excreted following an intake. After chemical isolation, the plutonium in urine samples may be determined by various methods including: alpha spectrometry (gas-flow proportional or surface-barrier detection), alpha counting (zinc sulfide or liquid scintillation counting), fission track counting, and mass spectrometry. Analytical procedures for in vitro measurement of plutonium and other radionuclides have been published (Volchok and dePlanque, 1983; Gautier, 1983).

Urine samples should be collected away from the plutonium facility to minimize cross-contamination. Samples should be collected in contamination-free containers; measures should be considered for minimizing plateout on walls of container surfaces (such as by addition of trace amounts of gold, oxalate, or nitric acid).

**Fecal Analysis.** Fecal analysis is a useful procedure for evaluating the excretion of plutonium and many other radioactive materials because more than half of the material deposited in the upper respiratory tract is cleared rapidly to the stomach and gastrointestinal (GI) tract.

The total fecal plus urinary elimination for the first few days after exposure, combined with in vivo counts that might be obtained, may provide the earliest and most accurate assessment of intake. Fecal samples taken during the second and third day after an inhalation incident are likely to provide the most useful data because the gastrointestinal hold-up time may vary from a few hours to a few days.

Fecal sampling is primarily a monitoring procedure for confirming and evaluating suspected intakes, but is used at some plutonium facilities for routine periodic monitoring as well. Workers may find fecal sampling unpleasant or objectionable, and laboratory technicians may also have aversion to fecal sample analysis. Some of these problems may be minimized if commercial fecal sample collection kits are used for convenient collection and handling of samples (Fisher et al., 1982). Collection kits also provide a means for collecting uncontaminated samples. Fecal samples may require additional sample preparation before analysis.

## **5.4 ESTABLISHING BIOASSAY FREQUENCY**

The bioassay measurement frequency should be based on 1) the potential risks of an intake occurring and 2) the sensitivity of a bioassay program to detecting potential intakes. The bioassay program sensitivity can be selected using specified intervals between measurements based on the MDD associated with an interval.

The rationale for the selected bioassay measurement frequency should also be documented. It is appropriate to evaluate the probability of intake and to modify the sampling frequency based on that probability.

The frequency of bioassay measurements should normally not be decreased because analytical results are below the detection level. The bioassay program should be maintained to confirm the proper functioning of the overall internal exposure control program and to document the absence of significant intakes of radionuclides.

### **5.4.1 Frequency Based on Program Sensitivity**

The minimum detectable dose concept refers to the potential dose associated with an MDA bioassay measurement at a given time interval post-intake. The pattern of retention of activity in the body, the MDA for a bioassay measurement technique, and the frequency with which that technique is applied define a quantity of intake that could go undetected by the bioassay program. An intake of such a magnitude would not be detected if it occurred immediately after a bioassay measurement and if it were eliminated from the body at such a rate that nothing was detected during the next scheduled measurement. The dose resulting from such an intake would be the MDD for that particular measurement technique and frequency.

Estimates of MDD in terms of CEDE should be documented for each measurement technique, MDA, and frequency. Retention functions specific to the various chemical forms and particle size distributions found in the facility should be used. Examples of MDD tabulations can be found in La Bone et al. (1993) and Carbaugh et al. (1994). In establishing MDD tables, it is important to consider dose contributions from all appropriate radionuclides in any mixture, rather than just the dose contribution from the bioassay indicator nuclide.

#### **5.4.2 Frequency Based on Potential Risk of Intake**

As discussed in Section 5.3.2, although plutonium workers are not generally considered to be at high risk of incurring intakes that might result in CEDEs of 100 mrem or more, any plutonium worker can be considered to have the potential for such an intake. However, having the potential for intake does not mean that they are likely to incur an intake.

Workers who have the highest potential risk for an intake are those most closely working with plutonium or plutonium-contaminated material. Typically, these workers are glove-box workers, maintenance workers, and operational health physics surveillance staff. These workers should be on a routine plutonium or americium bioassay program, including urinalysis and in vivo measurements. Such programs are relatively insensitive compared to the 100-mrem CEDE goal and are a safety net intended to catch intakes of significance relative to regulatory limits, rather than substantially lower administrative levels. Selection of bioassay frequency depends on the facility experience with potential intakes, the perceived likelihood of intake, and the MDD of a program. Annual urinalyses and in vivo chest counts are fairly typical. More frequent (e.g., semi-annual or quarterly) measurements may permit more timely review of workplace indicators in the event that an abnormal bioassay result is obtained, but do not necessarily mean a more sensitive program.

Plutonium facility decommissioning projects may present a different set of challenges for worker protection. In particular it is likely that clean up of areas will involve more plutonium that is not contained than is the case during normal operations. In addition, the workers involved may be relatively transient as the project progress through phases requiring different craft labor mixes. This being the case, more frequent bioassay may be necessary to provide good assurance that dose limits are not exceeded. As discussed in section 5.3.1, it is likely that program administrators will require a baseline measurement prior to the start of work and another at the termination of work. However, if the worker moves between tasks, it may be difficult to determine the source of an uptake without intermittent bioassay. In such cases, the use of breathing zone air samplers may be appropriate.

#### **5.4.3 Special Bioassay as Supplements to Routine Bioassay Programs**

Special bioassay programs for workers with known or suspected acute inhalation intakes of plutonium or other alpha-emitting radionuclides should include both urine and fecal sampling. Special bioassay measurements should be initiated for each employee in a contaminated work area when surface contamination is detected by routine surveillance if it is possible that the contamination resulted in a CEDE of 100 mrem or greater. Excreta samples should not be collected where they may be contaminated by external sources of plutonium. Ideally, total urine and feces should be collected for about a week following intake. This permits a sensitive assessment of potential intake and internal dose. Longer-term special samples collected at various times from a month to a year following intake can help to discriminate between ingestion, class W inhalation, and class Y inhalation. See Section 5.9 for indicator levels where special bioassay should be considered.

#### **5.4.4 Long-term Follow-up Bioassay Programs**

Following an intake a long-term follow-up bioassay program may be required for a worker to compare the actual excreta or in vivo results with those projected by the

evaluation. This is important to verify the accuracy of intake and dose assessments. The frequency and duration of a special program is dependent upon the projected values; it is suggested that as long as a worker continues to have detectable bioassay results, he or she should continue to be monitored. It is particularly important to have good baseline data and projections for individuals who return to plutonium work.

The ability of a bioassay program to distinguish between an established, elevated baseline and a new potential intake is important in the continued monitoring of workers once an intake has occurred. Because of statistical fluctuations in low-level plutonium and americium measurements, it can be very difficult to identify a new intake by routine bioassay if a worker has an elevated baseline.

## **5.5 ADMINISTRATION OF A BIOASSAY PROGRAM**

Administering a bioassay program requires that the policies, procedures, materials, support facilities, and staff be in place to enable a bioassay program to commence. Among the administrative items to address are the following:

- Management policy requiring participation in bioassay program by appropriate workers (may be part of an overall radiation protection policy)
- implementing procedures (e.g., criteria for who should participate, scheduling, sample kit instructions, sample kit issue/receipt, follow-up to unsuccessful sample or measurement attempts, data-handling)
- arrangements with appropriate analytical laboratories, including specifications of analysis sensitivity, processing times, reporting requirements, and quality assurance provisions



- onsite support facilities (e.g., sample kit storage locations, sample kit issue/collection stations, measurement laboratory facilities, equipment maintenance)
  
- staff selection, qualification, and training.

Recommendations for testing criteria for radiobioassay laboratories are in HPS N13.30 (HPS, 1996). These recommendations include calculational methods and performance criteria for bias, precision, and testing levels.

Some sites have established brief flyers or brochures describing their bioassay measurements. These may be distributed to workers during classroom training, upon notification of scheduled measurements, or at the time of the measurement or sample.

#### **5.5.1 In Vivo Monitoring**

The scheduling and measurement process for obtaining in vivo measurements is usually straightforward. Workers are scheduled for the measurements and results are available shortly after the measurement is completed. Counting times for in vivo  $^{241}\text{Am}$  measurements range from about 15 minutes to an hour or more, depending on the type of measurement and sensitivity required. The long counting times can impose limitations on the throughput of workers through a measurement facility, making scheduling an important issue. Procedures should be in place to assure that workers arrive for scheduled measurements and that follow-up occurs when a measurement is not completed or a worker fails to show.

Occasionally, workers are found who are claustrophobic when placed in an in vivo counter cells. Leaving the cell door partially open may help reduce some of the anxiety, but will also likely compromise the low background for which the system is designed.

Many workers want to know the results of their measurements. While a simple statement by the in vivo measurement technician may be adequate, a form letter stating that results were normal (or showed no detection of any of the nuclides of concern) can provide permanent verification. If results are not normal, a form letter can also be used to explain what happens next.

An important aspect of any in vivo measurement program is the calibration and verification testing of the measurement equipment. In vivo measurement results are highly dependent on the determination of a background result. Likewise, calibration using known activities in appropriate phantoms is also important. Phantoms are available commercially or by loan from the USDOE Phantom Library, operated by the Pacific Northwest Laboratory.<sup>(a)</sup>

### **5.5.2 Urine Sampling**

Urine sampling programs can be effectively administered using either workplace or home collection protocols. Workplace sampling protocols must assure that adequate precautions are taken to prevent external contamination of the sample by levels of activity well below the detection capabilities of friskers and workplace monitors. Home collection protocols have the advantage of being sufficiently removed from the workplace to render as essentially nonexistent the potential of very low-level contamination of the sample from external sources of plutonium. Avoidance of very minor external contamination of the samples is extremely important due to the dosimetric implications of plutonium in an in urine.

Large-volume urine samples are necessary for bioassay monitoring due to the very small urinary excretion rates. Ideally, 24-hour total samples would be preferred; however, such

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(a) For information on or to request loans from the USDOE Phantom Library, contact In an in Vivo Radioassay Research Facility, at the Pacific Northwest Laboratory, telephone (509) 376-6102.

samples often impose substantial inconvenience on workers, resulting in an in noncompliance with the instructions. As an alternative, total samples can be simulated by either time-collection protocols or volume normalization techniques.

One method of time-collection simulation (NCRP, 1987b; Sula et al., 1991) is to collect all urine voided from 1 hour before going to bed at night until 1 hour after rising in the morning for two consecutive nights. This technique has been reviewed with regard to uranium by Medley et al. (1994) and found to underestimate daily urine excretion by about 14%. Such a finding is not unexpected, since the time span defined by the protocol is likely to be about 18 to 22 hours for most people.

The volume normalization technique typically normalizes whatever volume is collected to the ICRP Reference Man daily urine excretion volume of 1400 mL. Reference Woman excretion (1000 mL/d) may be used for gender-specific programs. As a matter of practicality, routine monitoring programs do not usually use gender as a basis of routine data interpretation, particularly since results are anticipated to be nondetectable under normal conditions.

A third method calls for collection of a standard volume (e.g., 1 liter) irrespective of the time over which the sample is obtained. This method uses the standard volume as a screening tool only for routine monitoring. It does not attempt to relate measured routine excretion to intake, relying on well-defined and timely supplemental special bioassay to give true or simulated daily excretion rates.

The most common sample collection containers are 1-liter polyethylene bottles. Although glass bottles are also used, they pose additional risks of breakage. Wide-mouthed bottles are preferred for convenience and sanitation. The number of bottles included in the kit should be appropriate to the protocol; for a total 24-hour protocol as much as 3 liters can be expected. Special provisions, such as a funnel or transfer cup,

may improve the esthetics of sample collection and provide for added worker cooperation.

Some concerns can exist with length of sample storage before analysis. Storage may come from delays before batching samples in an in-house or due to transportation times to an offsite laboratory. The longer a sample stands, the more chemical and biological change it can undergo, typically manifesting itself as sedimentation and plateout on container walls. While samples can be preserved by acidification or freezing, good radiochemistry techniques should assure essentially complete recovery of any plateout or sediment. Samples sent offsite for analysis can be preserved with acid, but this method imposes hazardous material shipping requirements. Freezing samples can preserve them, but plateout and sedimentation upon thawing should still be expected.

Precautions are necessary if a lab uses an aliquot for analysis and extrapolates the aliquot result to the total sample. The aliquoting procedure should be tested using spiked samples to assure that it is representative.

A quality control (QC) verification program should exist for laboratory analyses, including use of known blank samples and samples spiked with known quantities of radioactivity. Ideally, the samples should not be distinguishable by the analytical laboratory from actual worker samples. The number of QC verification samples may range from 5% to 15% of the total samples processed by a large-volume program; a small program focused on submittal of special samples following suspected intakes may have a much higher percentage of controls. An additional QC provision may be to request the analytical lab to provide results of their in an in-house QC results for independent review.

There are no standard or regulatory requirements for bioassay sample chain-of-custody provisions, nor has there been consensus on their need. Tampering with samples has not been a widely reported or suspected problem. Site-specific chain-of-custody requirements should be based on balancing the need with the resources required to

implement them. Some sites have no chain-of-custody requirements associated with bioassay sample collection. At other sites, a simple seal placed on a sample container following collection by the subject worker is an effective means of providing a small degree of chain-of-custody. At the more complex level would be strict accountability requiring signature of issue, certification of collection, and signature of submittal.

Procedures describing details of the bioassay program should be documented. These procedures should include a description of sample collection, analysis, calibration techniques, QC, biokinetic modeling, and dose calculational methods used.

### **5.5.3 Fecal Sampling**

A fecal sampling program must be designed to ensure worker cooperation, whether collecting samples at home or in an in the workplace. Since the frequency of fecal voiding varies greatly from person to person, the sample collection program must be adaptable. Flexibility in an in sample dates is important. It is suggested that when a fecal sample is required, the worker be provided with a kit and instructed to collect the sample, noting the date and time of voiding on the sample label. This practice can reduce the likelihood of unsuccessful samples. If multiple samples are required (for example, to collect the total early fecal clearance following an acute inhalation exposure), the worker may be given several kits and told to collect the next several voidings, noting the date and time of each.

Since the total fecal voiding should be collected, thought must be given to the kit provided. Fecal sampling kits can be obtained from medical supply companies or designed by the site. A typical kit might include a large plastic zipper-closure bag to hold the sample, placed inside a 1- to 2-liter collection bucket with a tight-fitting lid. The bucket and bag can be held in an in place under a toilet seat by a trapezoid-shaped bracket with a hole through it sized to hold the bucket. After sample collection, the

zipper bag is sealed, the lid is snapped tight on the bucket, and the bucket placed in an in a cardboard box.

Following collection, the sample handling, control, analytical, and QC provisions are similar to those described above for urine samples. One particular concern for fecal analysis is the potential difficulty of dissolving class Y plutonium in an in the fecal matrix. While nitric acid dissolution may be adequate, enhanced digestion using hydrofluoric acid may be preferred.

## **5.6 MODELING THE BEHAVIOR OF PLUTONIUM IN AN IN THE BODY**

A key issue to plutonium dosimetry is the modeling of how the material behaves in an in the body. Some of the standard models are described below, with additional discussion on the biological behavior given in an in Section 2.4. It is important that an internal dosimetry program establish and document the routine models and assumptions used for dosimetry. Computer codes typically incorporate standard models but may allow the flexibility to alter parameters. When altered on an individual-specific basis, the revised models need to be addressed in an in the pertinent case evaluations or the technical basis.

### **5.6.1 Respiratory Tract**

The respiratory tract model of ICRP Publication 30 (1979 and 1988b) is commonly used for evaluating inhalation intakes of radioactivity. The model has been widely published and included in an in reference books (e.g., Cember, 1996; Shleien, 1983) and internal dosimetry computer codes, hence it is not reproduced here.

Like all models, the ICRP respiratory tract model represents anticipated behavior. Once an exposure has occurred and actual data become available, deviations from the model in an in light of the data are appropriate.

In an in practice, the model has proved extremely valuable for calculating derived investigation levels and estimating intakes from bioassay data, using standard D, W, and Y classes of material. Model interpretation becomes more subjective when extensive data become available. Carbaugh et al. (1991) and La Bone et al. (1992) have provided excellent examples of two cases where the standard lung model assumptions did not fit the data.

Most internal dosimetry computer codes allow adjustment of particle size and selection of solubility classes. Some codes also permit detailed adjustment of the model's individual compartment parameters; with these codes, it may be possible to arrive at various subjective interpretations to explain the same data. When adjustments are made to the standard assumptions, it is important to explain what those adjustments are and why they were made.

#### **5.6.2 Gastrointestinal Tract**

The gastrointestinal tract model of ICRP Publication 30 (1979 and 1988b) is also widely promulgated and used for evaluating ingestion intakes and as well as being coupled to the respiratory tract for inhalation intakes. The model is particularly subject to individual variations in an in fecal voiding frequency, so judgment must be used in an in its application to human data.

A key parameter of the model for internal dosimetry is the  $f_i$  factor for absorption to blood of material in an in the small intestine. The  $f_i$  factor varies from  $10^{-5}$  for

plutonium oxides to  $10^{-4}$  for plutonium nitrates and to  $10^{-3}$  for other compounds and americium.

### **5.6.3 Systemic Retention and Excretion of Plutonium**

Standard models for the systemic retention of plutonium are commonly used for internal dosimetry because in an in vivo detection of plutonium within the individual systemic compartments is not usually possible. Three models proposed by the ICRP over a 10-year period are described in an in Section 2.4.2 of this document. Each of them has had a wide application, and ICRP has suggested that results derived using one model do not need to be rederived for compliance purposes using the newest model. Studies by the U.S. Transuranium Registry and summarized by Kathren (1994) have indicated that alternate compartments and clearance half-times may be more appropriate.

For convention, this document will use the ICRP 30, Part 4 (1988b) systemic retention parameters for plutonium internal dosimetry. That model assumes that for plutonium reaching the blood, 45% is deposited on bone surfaces from which it clears with a 50-year half-time, 45% is deposited in an in the liver with a 20-year clearance half-time, and a very small fraction ( $3.5 \times 10^{-4}$  for males and  $1.1 \times 10^{-4}$  for females) is permanently retained in an in gonadal tissue. The remaining 10% goes uniformly to all other tissues and direct excretion.

Excretion models for plutonium include the empirical models of Langham (1956) and Langham et al. (1980), Durbin (1972), Jones (1985), and Tancock and Taylor (1993), as well as study models such as Leggett (1984). This technical document does not take a position on the "best" model. Site choices of dosimetry tools such as reference tabulations (Lessard et al., 1987; ICRP, 1988a) and computer codes may dictate one model over another. The choice of model and explanation of its selection are among the technical bases of the site internal dosimetry program.





#### **5.6.4 Natural Plutonium Balance in an in Man**

Although plutonium can be found in an in members of the general public as a result of worldwide fallout from atomic weapons detonations, the levels are quite small. A summary of the literature can be found in an in ICRP Publication 48 (1986). Data from McInroy et al. (1979, 1981) suggests that median body burdens of plutonium in an in the U.S. population peaked at about 12 pCi during the 1960s and declined to about 2 pCi by 1977. Tissue concentration data from Nelson et al. (1993) can be used to calculate a median body burden in an in the early 1970s of 3 to 4 pCi.

These body burdens imply that urinary or fecal excretion associated with worldwide fallout will not be detectable by routinely available bioassay procedures. Consequently, it is reasonable to assume that any bioassay detection by a worker-monitoring problem is likely to be attributable to occupational exposure.

#### **5.6.5 Mother-to-Fetus Transfer**

The embryo/fetus is included as part of the 10% of the systemic uptake that is uniformly distributed in an in all "other" soft tissues except the liver and gonads. Methods for evaluating embryo/fetal uptake and dose have been described by Sikov et al. in an in NUREG/CR-5631 (1992) and its 1993 addendum (Sikov and Hui, 1993). For uptakes occurring during the first 2 months of pregnancy, the activity in an in the embryo/fetus is assumed to have the same concentration as in an in the mother's "other soft tissue." For later uptakes, the embryo/fetal concentration gradually increases relative to the maternal concentration, but is assumed to remain uniformly distributed in an in the embryo/fetus. At 3 months, the embryo/fetal concentration is 1-1/2 times the mother's "other" soft tissues concentration. At 6 months, it is twice the mother's, and at 8 months it is thrice the maternal "other" concentration. Following transfer to the embryo/fetus, activity is assumed to remain, without clearance, until birth.

The Nuclear Regulatory Commission has developed simplified methods for assessing the gestation period dose to an embryo/fetus in an in Regulatory Guide 8.36 (NRC, 1992). Application of these methods shows that very large maternal intakes of plutonium or americium are required to produce uptakes that would deliver 500 mrem, or even 50 mrem to the embryo/fetus. The NUREG/CR-5631 Addendum (Sikov and Hui, 1993) notes that maternal inhalation intakes of nominally 100 times the annual limit on intake (ALI) are required to give a 50-mrem embryo/fetal dose. For ingestion intakes, a 1,000 ALI maternal intake of plutonium is required to give a 50-mrem dose to the embryo/fetus. Thus, providing adequate radiation protection to limit maternal intake of plutonium and americium to the occupational limits will adequately provide for the protection of the embryo/fetus.

## **5.7 INTERPRETATION OF BIOASSAY RESULTS**

Bioassay measurements detecting plutonium or americium in an in workers can be initially interpreted as indicating that occupational intakes may have occurred. Standard bioassay procedures are not sufficiently sensitive to detect the worldwide environmental background levels in an in vivo or in an in excreta. Since most plutonium and americium bioassay measurement procedures include counting for radioactivity as the final step in an in the measurement process, they are subject to the statistics associated with the counting process.

Two key questions associated with bioassay data are 1) When does a sample result indicate the presence of something (i.e., when is the analyte detected)? and 2) What is the overall capability of the bioassay method for continual assurance of detection of the analyte?

The decision level,  $L_c$  (also called the critical level for detection), is the level for a given measurement that indicates the likely presence of the analyte. The  $L_c$  is dependent on the probability of obtaining false positive results (type I, or alpha, error) that is acceptable to the program. A 5% probability of false-positive results is a common design parameter of measurement programs, implying that for a large number of measurements, 5% of the time results

will be indicated as positive when in an in fact there is no activity present. The  $L_c$  is calculated from results of analyses of blank samples. Once a measurement is performed, it is appropriate to compare it with the  $L_c$  to determine whether or not the result is “positive” (i.e., the analyte is detected).

The MDA is the level at which continued assurance of detection can be provided. The MDA is a function of the probabilities of both false positive and false negative (type II, or beta) errors and is typically based on a 5% probability for each kind of error. The MDA is also determined from analysis of blank samples, but is substantially higher than the  $L_c$ . The MDA is appropriate for use in an in designing bioassay programs and as the basis for estimating minimum detectable intakes and doses as indicators of program sensitivity. The MDA should not be used as a comparison with actual measurements to determine whether or not activity is present (i.e., <MDA is not an appropriate use of the concept).

Methods for calculating both  $L_c$  and MDA are given in an in HPS N13.30. (HPS, 1996).

As an alternative to the  $L_c$  and MDA of classical statistics, Miller et al. (1993) propose the use of Bayesian statistical methods for evaluating bioassay data.

General follow-up actions to abnormal bioassay measurements should include data checks, timely verification measurements, work history reviews, and performance of special in an in vivo measurements or excreta sample analyses for intake and dose assessments.

### **5.7.1 In an in Vivo Count Results**

In an in vivo plutonium or americium measurements are generally relatively insensitive with regard to levels of occupational exposure concern. This applies particularly to routine chest or lung counting, skeleton counting, and liver counting. For that reason, any detection of plutonium or americium should be investigated. The investigation should address the validity of the measurement by reviewing the spectrum and its associated background subtraction. These reviews are particularly important if the result is near the  $L_c$ . Follow-up to a positive result should include a confirming measurement. Ideally, this should be an immediate (same day) recount of equal or higher sensitivity. The farther removed in an in time a verification measurement is from the original measurement, the more important it becomes to factor in an in potential lung clearance in an in comparing the two measurements. A follow-up measurement taken 30 days after an initial high-routine may not be capable of providing verification if the material of concern exhibits class W behavior.

Chest-wall thickness has a significant impact on chest counting. Corrections are commonly made using a height-to-weight ratio or ultrasonic methods (Kruchten and Anderson, 1990).

Corrections may be required to address apparent detection in an in one tissue resulting from photon crossfire from another tissue. For example, chest counting is performed primarily to estimate activity in an in the lung. Yet, there is substantial bone over the lungs (rib cage, sternum) and behind the lungs (vertebrae). Plutonium and americium are both bone-seeking radionuclides which will deposit on those bone surfaces and can interfere with chest counting. It is possible for a person having a systemic burden of plutonium from a wound in an in the finger to manifest a positive chest count from material translocated to the skeleton, axillary lymph nodes, or liver (Carbaugh et al., 1989; Graham and Kirkham, 1983; Jeffries and Gunston, 1986). Interpreting such a chest count as a lung burden can render dose estimates somewhat inaccurate.

When comparing in vivo measurements made over many years, it is important to make sure that the measurements are, in fact, comparable. One consideration is to make sure that corrections have been consistently applied to all similar measurements. It is not unusual for measurement systems to be replaced or to change the algorithms used for calculating results over time. Step changes in data can occur and should be addressed in monitoring long-term detectable trends (Carbaugh et al., 1988).

In vivo wound counting for plutonium or americium is usually one facet of special bioassay following a wound. While a portable alpha survey meter may show if surface contamination is present at the wound site or contamination of the wounding object, alpha detectors are not capable of measuring imbedded activity or activity masked by blood or serum. Thus, plutonium and americium facilities should have available a wound counter utilizing a thin sodium iodide or semi-conductor (e.g., planar germanium) detector. Such detectors are capable of measuring the low-energy photons emitted from plutonium and americium. The ability to accurately quantify wound activity is highly variable, depending on the calibration of the equipment and how deeply imbedded material is in the wound. If the object causing a wound and blood smears taken at the time of a wound show no detectable activity, then a wound count also showing no detectable activity is probably sufficient to rule out an intake. If the wounding object or the blood smears show detectable activity, special urine samples should be obtained regardless of the wound count result. In this latter circumstance, lack of detectable activity on a wound count could be attributable to deeply imbedded material at the wound site or to rapid transportation of material from the wound to the systemic compartment.

Ingrowth of  $^{241}\text{Am}$  from  $^{241}\text{Pu}$  in plutonium mixtures can also significantly impact in vivo data interpretation. Rather than decreasing with time,  $^{241}\text{Am}$  results can increase without additional intake. This circumstance is particularly likely if dealing with residual activity bound up in wound sites, but may also be observed by in vivo chest or skeleton counting. A method to evaluate  $^{241}\text{Am}$  ingrowth is described in Section 5.8.4.

### 5.7.2 Urine Sample Results

Detection of plutonium or americium activity in an in a routine or special urine sample using commonly available radiochemical measurement techniques should be investigated as a potential intake. A data review should be made to assure that the sample result was correctly determined, and batch QC sample data should be verified.

If the result is near the  $L_c$ , it is possible that statistical fluctuation of the measurement process could account for the apparent detection. Recounting the final sample preparation once or twice can be a helpful technique to verify a result or classify it as a false-positive. If the first recount also detects the analyte, it can be concluded that the sample does contain the analyte (the likelihood of two consecutive false positives at a 5% type I error per measurement is 0.0025, or 0.25%.) If the first recount does not detect the analyte, a second recount can be performed as a tie-breaker.

An investigation should be initiated for any abnormal plutonium or americium urinalysis result. “Abnormal” for a person with no prior history of intake should be interpreted as any detectable activity.

Once an intake is confirmed, sufficient samples must be obtained to establish a reasonably anticipated baseline against which future measurements can be compared. This is important both to provide future verification of the accuracy of the assessment and to identify potential additional intakes.

The statistical fluctuation of low-level measurements can be particularly troublesome for long-term excretion patterns. Factors of 2 can be easily expected due to day-to-day variability and imprecise adherence by the worker to urine collection protocols.

### **5.7.3 Fecal Sample Results**

Fecal samples are much more sensitive to detection of intakes than are urine samples and, consequently, are an important part of follow-up bioassay monitoring for potential intakes initially identified by workplace indications. Pitfalls to the data interpretation include highly variable individual fecal voiding patterns, ranging from more than one per day to one every few days. This makes it extremely important to know what time interval is represented by a collected fecal sample. While normalizing a single set of fecal data to reference man daily excretion rate can be done, it is not likely to improve the quality of assessment.

The preferred fecal sampling protocol following an intake is to collect all the early fecal clearance (meaning total feces for the first five-to-seven days). This method will allow a good estimation of inhalation or ingestion intake, but does not readily permit discrimination of inhalation from ingestion, or identify whether inhaled material exhibits class D, W, or Y clearance patterns. For optimum interpretation, total fecal collection should be interpreted in an in light of early urine and in an in vivo data for preliminary estimates. The urine data is likely to be particularly valuable in an in conjunction with fecal data to classify an intake as class W or Y. Longer-term follow-up fecal samples at nominally 30, 60, and 90 days post-intake should substantially improve the classification of material as class W or Y.

Fecal sampling can also be applied to monitor excretion at long times post-intake. One caveat in an in such sampling is that a worker still active in an in a plutonium facility may be incurring very minor chronic exposure, which can significantly interfere with long-term interpretation of acute exposure data. Bihl et al. (1993) have discussed experience with a routine fecal sampling program.



**5.7.4 Use of Air Sample Data in an Internal Dosimetry**

Results of air sampling and continuous air monitoring implying more than 40 DAC-hours exposure should be used to initiate special bioassay to assess intakes of plutonium.

Although bioassay data are the preferred method for assessing intakes and internal doses, air sample data can be used for assessing internal doses if bioassay data are unavailable or determined to be inadequate or nonrepresentative. Air sample data can be used to calculate an exposure to airborne material either in terms of DAC-hours or potential radioactivity intake as follows:

$$\text{DAC} - \text{hours} = \frac{\text{Air Concentration}}{\text{DAC}} * \text{Duration (hours)} \quad (5.2)$$

$$\text{Intake} = \text{Air Concentration} * \text{Breathing Rate} * \text{Time} \quad (5.3)$$

DAC = The airborne concentration for radionuclides listed in an in Appendix A of 10 CFR 835, taking into consideration the Clearance Class (D/W/Y) expressed in  $\mu\text{Ci/mL}$  or  $\text{Bq/m}^3$

Air concentration = airborne radioactivity in units of  $\mu\text{Ci/mL}$  or  $\text{Bq/m}^3$

If air sample results are representative of air breathed by individuals, then doses can be calculated using the 5-rem stochastic limit for CEDE ( $H_{E,50}$ ) or the 50-rem nonstochastic limit for committed tissue dose equivalent ( $H_{T,50}$ ) and the respective stochastic or nonstochastic DAC or ALI conversion factor, as shown below:

$$H_{50} = (\text{DAC} - \text{hours}) * \frac{\text{Dose Limit}}{2000 \text{ DAC} - \text{hours}} \quad (5.4)$$

$$H_{50} = \text{Intake} * \frac{\text{Dose Limit}}{\text{ALI}} \quad (5.5)$$

If respiratory protection is worn by workers, the appropriate respirator protection factor may be applied to the above calculations (i.e., dividing the calculated result by the protection factor.)

General air sampling programs should be augmented by breathing zone sampling when air concentrations to which individuals are exposed might be highly variable. Breathing zone sampling may include both fixed-location and personal (lapel) air samplers.

Personal air samples are more likely to be representative of actual exposure conditions than are samples collected at fixed locations, and can be particularly useful for assessing potential intakes involving short-term exposure to well-monitored air concentrations.

## 5.8 DOSE ASSESSMENT

Dose assessment involves collecting and analyzing information concerning a potential intake and developing a conclusion regarding the magnitude of intake and its associated committed dose equivalents. Dose assessments are conducted by investigating the nature of a potential intake and by analyzing bioassay measurement results or other pertinent data. Biokinetic models are used in an in conjunction with bioassay data to evaluate the intake, uptake, and retention of plutonium in an in the organs and tissues of the body. Intake estimates can then be used to calculate committed effective and organ dose equivalents. It is essential that good professional judgement be used in an in evaluating potential intakes and assessing internal doses. Carbaugh (1994) has identified a number of considerations for dose assessments.

Computer codes are commonly used for assessment of intakes, dose calculation, and bioassay or body content projections. La Bone (1994a) has provided an overview of what should be considered in selecting a computer code, as well as descriptions of a number of internal dosimetry codes available in 1994. Internal dosimetry code users should understand how the code works and be aware of its limitations. Computer codes merely provide the logical result of the input they are given. Use of a particular computer code does not necessarily mean a dose estimate is correct.

As used in this section, the definition of "intake" is the total quantity of radioactive material taken into the body. Not all material taken into the body is retained. For example, in an inhalation intake, the ICRP Publication 30 respiratory tract model predicts that, for 1- $\mu$ m particles, 63% of the intake will be deposited in the respiratory tract; the other 37% is immediately exhaled (ICRP, 1979). For a wound intake, material may be initially deposited at the wound site. Once the material has been deposited, it can be taken up into systemic circulation either as an instantaneous process (e.g., direct intravenous injection of a dissolved compound) or gradually (e.g., slow absorption from a wound site or the pulmonary region of the lung). Both the instantaneous and slow absorption processes are often referred to as uptake to the systemic transfer compartment (i.e., blood). Once material has been absorbed by the blood, it can be translocated to the various systemic organs and tissues.

An understanding of this terminology is important to review of historical cases. In the past sites reported internal doses not as dose equivalent estimates but as an uptake (or projected uptake) expressed as a percentage of a maximum permissible body burden (MPBB). The standard tabulated values for MPBBs were those in ICRP Publication 2 (ICRP, 1959). Many archived historical records may have used this approach. DOE Order 5480.11 (superseded), required calculation of dose equivalent. Now, 10 CFR 835 (DOE, 1993c), has codified the calculation of intakes and committed doses.

**5.8.1 Methods of Estimating Intake**

There are several published methods for estimating intake from bioassay data (Skrable et al., 1994a; Streng et al., 1992; ICRP, 1988b; King, 1987; Johnson and Carver, 1981). These methods each employ an idealized mathematical model of the human body showing how materials are retained in an in and excreted from the body over time following the intake. An intake retention function (IRF) is a simplified mathematical description of the complex biokinetics of a radioactive material in an in the human body. These functions are used to predict the fraction of an intake that will be present in an in any compartment of the body, including excreta, at any time post-intake. Intake retention functions incorporate an uptake retention model that relates uptake to bioassay data and a feed model that relates intake to uptake and bioassay data. ICRP Publication 54 (1988a) and Lessard et al. (1987) have published compilations of IRFs. Selected IRFs calculated using the GENMOD Computer Code (incorporating the Jones excretion function) for the lung, urine, and fecal excretion are show in an in Tables 5.6 for class W and Y forms of  $^{239}\text{Pu}$ . These functions would be similar in an in value to those for other long-lived forms isotopes of Pu.

$$Q_t = \text{Intake} * \text{IRF} (Q_t) \quad (5.6)$$

In an in its simplest form, a compartment content at any time post-intake ( $Q_t$ ) can be expressed as the product of intake multiplied by the intake retention function value for compartment Q at time t post-intake, or:

Results predicted by the model can then be compared with the observed bioassay data. Such results are often referred to as expectation values.

**Table 5.6.** Intake Retention Fractions<sup>(a)</sup> for <sup>239</sup>Pu

Days Post-Intake	Class W			Class Y		
	Lung	Urine	Feces	Lung	Urine	Feces
1	2.16E-01	2.36E-04	1.05E-01	2.14E-01	1.28E-05	1.27E-01
2	1.78E-01	1.61E-04	1.34E-01	1.81E-01	8.71E-06	1.56E-01
7	1.39E-01	3.29E-05	6.41E-03	1.50E-01	1.69E-06	5.20E-03
14	1.26E-01	2.18E-05	1.23E-03	1.48E-01	1.09E-06	1.73E-04
30	1.03E-01	1.73E-05	9.58E-04	1.45E-01	8.60E-07	1.34E-04
60	7.01E-02	1.34E-05	6.34E-04	1.41E-01	7.15E-07	1.29E-04
90	4.77E-02	1.15E-05	4.21E-04	1.37E-01	6.89E-07	1.23E-04
180	1.50E-02	8.31E-06	1.26E-04	1.25E-01	7.19E-07	1.09E-04
365	1.36E-03	4.84E-06	1.39E-05	1.04E-01	7.97E-07	8.46E-05
730	1.09E-05	2.45E-06	2.50E-06	7.31E-02	8.85E-07	5.14E-05
1825	0.00E+00	1.63E-06	1.63E-06	2.90E-02	8.82E-07	1.19E-05
3650	0.00E+00	1.54E-06	1.54E-06	9.60E-03	7.38E-07	1.36E-06
7300	0.00E+00	1.38E-06	1.38E-06	4.16E-03	6.09E-07	6.15E-07
18250	0.00E+00	1.00E-06	1.00E-06	3.70E-03	4.38E-07	4.39E-07

(a) Calculated using GENMOD Computer Code Version 3.0.3(2), assuming 1- $\mu$ m AMAD aerosol.

Simple algebraic manipulation of the model allows calculation of intake from the compartment content at time t, as shown below:

$$\text{Intake} = \frac{Q_t}{\text{IRF}(Q_t)} \quad (5.7)$$

When multiple data points are available for a compartment, the intake can be estimated using an unweighted or weighted least-squares fitting procedure, as described by Skrable et al. (1994b) and Strenge et al. (1992) or as can be found in an in most statistics textbooks. As an alternative, data can be fit by eye to a graphical plot; however, the apparent fit can be misleading if data has been logarithmically transformed.

Intake can also be estimated from air sample data, as described in an in Section 5.7.4. This method is appropriate if bioassay data are not available or insufficiently sensitive. Intake estimates based on air samples and bioassay data are also appropriate as a check on each other. Valid bioassay data showing detectable results should be given preference over intake estimates based on air sample results.

### **5.8.2 Alternate Methods of Intake Assessment**

Historically, intake as described in an in the foregoing section was not always calculated when assessing plutonium exposures. Estimates of uptake using methods similar to Langham (1956), Healy (1957), or Lawrence (1987) focused on assessing the magnitude of radioactivity retained in an in the body, rather than intake (which includes material not retained and of no dosimetric significance). These methods were (and are) dosimetrically sound in an in so far as estimates of deposition and uptake are concerned, but do not meet the current regulatory requirement of 10 CFR 835 (DOE, 1993c) to calculate intake.

### **5.8.3 Estimating Effective Dose Equivalent from Intakes of Plutonium**

The committed dose equivalent resulting from an intake of plutonium may be calculated by multiplying the estimated intake (I) by an appropriate dose conversion factor (DCF):

$$H_{50} = I * DCF \quad (5.8)$$

Dose conversion factors can be obtained from tabulated data in an in Federal Guidance Report No. 11 (EPA, 1988b), ICRP 30, Part 4 (1988b), in an in the Supplement to Part 1 of ICRP 30 (ICRP 1979), or calculated directly using computer programs. Substituting the ICRP 48 (1986) model parameters of 50% skeleton and 30% liver translocation for the assumptions in an in ICRP 30, Parts 1 or 4, has little impact on the  $H_{E,50}$  per unit intake, but does alter the committed organ dose equivalent per unit intake. Such substitution of models is acceptable, provided that the model is documented and consistently applied.

Values for simplified dose conversion factors can be obtained by dividing a dose limit by the corresponding value for the ALI. A caution must be observed with this approach: not all tabulated values of ALIs are the same. The ALIs are commonly rounded in most tabulations to one significant figure (e.g., as in ICRP Publication 30 and Federal Guidance Report No. 11). Substantial variation can occur as a result of units conversion. For example, Federal Guidance Report No. 11 lists the ALI for  $^{239}\text{Pu}$  class Y inhalation as both  $6 \times 10^{-4}$  MBq (600 Bq) and 0.006  $\mu\text{Ci}$  (740 Bq). Such rounding errors can introduce significant discrepancies in dosimetry calculations. This method also raises a question about which ALI should be used if compliance monitoring is being based on comparison with secondary limits, such as the ALI rather than the primary dose limits.

Where individual-specific data are available, the models should be adjusted. However, the general lack of capability to monitor organ-specific retention for plutonium (i.e., content and clearance half-times) makes the use of default models most practical.

Ideally, one should obtain as much bioassay information as possible to determine the intake and track the retention of plutonium in the body to reduce the uncertainty associated with the daily variation in the measurements. A regression analysis should be used to fit the measurement values for estimating the initial intake and clearance half-times.

#### **5.8.4 Evaluating $^{241}\text{Am}$ Ingrowth In an in Vivo**

Ingrowth of  $^{241}\text{Am}$  from  $^{241}\text{Pu}$  can significantly impact bioassay monitoring projections. Unless accounted for, it can lead to suspicion of new intakes, or underestimation of clearance rates. The amount of  $^{241}\text{Pu}$  present in a plutonium mixture depends on the irradiation history and time since irradiation. Freshly processed mixtures containing 6% by weight of  $^{240}\text{Pu}$  may contain about 0.5% by weight of  $^{241}\text{Pu}$  and a 12%  $^{240}\text{Pu}$  mixture may contain 3%  $^{241}\text{Pu}$ . Commercial spent fuel can be much higher. The ingrowth of

$^{241}\text{Am}$  occurs following a plutonium intake over a period of years. Less transportable (Class Y) forms of plutonium may have  $^{241}\text{Am}$  ingrowth which gradually becomes detectable. An extreme case of this was demonstrated in an in a well-documented Hanford plutonium-oxide exposure which exhibited a factor-of-2 increase in an in  $^{241}\text{Am}$  lung content in an in the 3000 days following intake (Carbaugh et al., 1991). Such an increase could not be explained using the standard 500-day class Y lung clearance half-time; finally, a 17-year biological clearance half-time was estimated. The subsequent committed effective dose equivalent was estimated to be a factor of 3 higher than if the standard 500-day half-time had been used. Similar difficulties have occurred with initial detection of  $^{241}\text{Am}$  by routine in an in vivo chest counting or in an in long-term monitoring of residual wound content.

While many available internal dosimetry computer codes will calculate the projected  $^{241}\text{Am}$  lung content following an intake (accounting for ingrowth in an in the process), none of the current codes will do curve-fitting from long-term data and at the same time adjust the data for ingrowth. Therefore, the following simplistic method was developed to assess that data.

An estimate of the  $^{241}\text{Am}$  ingrowth can be made by assuming that, at the time of intake ( $t = 0$ ), all the material that will compose the long-term component is deposited in an in a single compartment and that the rate of transfer of material from the compartment at any subsequent time  $t$  is proportional to the quantity of material remaining in an in the compartment (i.e., simple exponential transport kinetics). The following equation will then describe the buildup of  $^{241}\text{Am}$  in an in that compartment following an initial deposition of  $^{241}\text{Pu}$  and  $^{241}\text{Am}$  and a given or assumed effective clearance rate:



$$A_{t,Am} = \lambda_{r,Am} \frac{A_{0,Pu}}{k_{e,Am} - k_{e,Pu}} (e^{-k_{e,Pu}t} - e^{-k_{e,Am}t}) + A_{0,Am} e^{-k_{e,Am}t} \quad (5.9)$$

where  $A_{t,Am}$  = activity of  $^{241}\text{Am}$  at time  $t$

$\lambda_{r,Am}$  = radiological decay constant for  $^{241}\text{Am}$

$A_{0,Pu}$  = activity of  $^{241}\text{Pu}$  at time 0

$k_{e,Am}$  = effective clearance rate of  $^{241}\text{Am}$

$k_{e,Pu}$  = effective clearance rate of  $^{241}\text{Pu}$

$A_{0,Am}$  = activity of  $^{241}\text{Am}$  at time 0

$t$  = elapsed time

The effective clearance rate ( $k_e$ ) of any nuclide is the sum of the radiological decay constant ( $\lambda_r$ ) and the biological clearance rate ( $\lambda_{bio}$ ). By assuming that the biological

clearance rate is constant for both parent and progeny nuclides, the equation reduces to three unknowns: the initial amount of parent, the initial amount of progeny, and the biological clearance rate. These unknowns can be dealt with by assuming a standard isotopic composition at the time of intake and then solving the equation for a biological clearance rate using an iterative process until the calculated result matches the observed result at a given time  $t$ . A computer or calculator algorithm can eliminate the need for lengthy hand calculations.

Once an optimum combination of isotopic compositions and biological clearance rate is found, internal dosimetry codes or hand calculations can be used to estimate organ and effective doses. As a check on the results, standard computer codes can be used in a bioassay projection mode to project the  $^{241}\text{Am}$  content based on the estimated intake and biological clearance rate.

## 5.9 INDICATOR AND ACTION LEVELS

Indicator and action levels are essential to operation of a routine internal dosimetry program. Because a wide range of levels can be defined by various facilities and organizations, this document does not attempt to prescribe particular level titles. As used in an in this document, indicator and action levels are simply workplace or bioassay measurements, or associated calculated doses, at which specific actions occur.

Indicator levels based on workplace indicators for reacting to a potential intake are suggested in an in Table 5.7. The intent of these indicator levels is to provide guidance for field response to any potential intake of radioactive material with a potential for a dose commitment that is  $>100$ -mrem CEDE. It is suggested that when these levels are reached, appropriate management members of the health physics and operations organizations be informed. See Section 5.4.3 for guidance on special bioassay. Table 5.8 suggests notification levels to the occupational medicine physician for possible early medical intervention in an in an internal contamination event. These tables, derived from Carbaugh et al. (1994), are based on general considerations and significant experience with past intakes of radioactive material and, because they are based on field measurements, do not correspond with any exact dose commitment to the worker.

The decision to administer treatment and the treatment protocol are solely the responsibilities of the physician in an in charge. The basic principle is that the proposed intervention should do more good than harm (Gerber and Thomas, 1992).

Guidelines for the medical intervention of a radionuclide intake can be found in an in several publications. NCRP Report No. 65 (NCRP, 1980) and the joint publication of the Commission on European Communities (CEC) and the DOE Guidebook for the Treatment of Accidental Internal Radionuclide Contamination of Workers (Gerber and Thomas, 1992) both contain detailed guidance in an in intervention and medical procedures useful in an in mitigating radiation overexposures. The ICRP recommends in an in Publication 60 (1991b) a limit of 2-rem/y (20-mSv/y) on effective dose. Thus, the ALIs found in an in ICRP Publication 61 (1991a) and

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

used in an in the CEC/DOE Guidebook noted above are those which would provide a CEDE of 2-rem/y instead of current U.S. regulations of 5-rem/y.

**Table 5.7.** Plutonium or  $^{241}\text{Am}$  Indicator Levels for Internal Dosimetry Evaluation

Indicator	Notification Level
Nasal or mouth smears	Detectable activity
Facial contamination	200 dpm
(direct measurement)	
Skin breaks or blood smears	Any skin break while handling material other than sealed sources
Head, neck contamination	2,000 dpm
Contamination in an in respirator	Detectable activity inside respirator after use
Hands, forearms, clothing	10,000 dpm
contamination <sup>(a)</sup>	
Airborne radioactivity	Acute intake equivalent to 40 DAC-hours after accounting for respiratory protection factor

(a) Clothing contamination levels apply to exposure without respiratory protection, such as on inner coveralls or personal clothing.

**Table 5.8.** Plutonium or  $^{241}\text{Am}$  Contamination Levels for Notification of Occupational Medicine Physician

Indicator	Medical Notification Level, dpm
Nasal or mouth smears	1,000
Facial contamination	25,000
Skin breaks or wounds	100

Guidance in an in the CEC/DOE Guidebook can be summarized as follows:

- When the estimated intake is below one ALI, treatment should not be considered.
- When the estimated intake is between 1 and 10 times the ALI, treatment should be considered. Under these situations, short-term administration will usually be appropriate, except for intake of materials poorly transported from the lung (class Y).

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- When the estimated intake exceeds 10 times the ALI, then extended or protracted treatment should be implemented, except for materials poorly transported from the lung.
- For poorly transported material in an in the lung, lung lavage is the only recommended treatment, and it is only a consideration for intakes exceeding 100 times the ALI.

Because the dose associated with the ALI in an in the CEC/DOE Guidebook is 2-rem CEDE and because the upper administrative level recommended by the RCM is 2 rem, intervention levels of 2 rem and 20 rem might be used for guidance in an in the manner presented in an in the CEC/DOE Guidebook:

- When the CEDE estimated intake is below 2 rem, treatment is not generally recommended.
- When the CEDE for an estimated intake is between 2 rem and 20 rem, treatment should be considered. Under these situations, short-term administration will usually be appropriate.
- When the CEDE equivalent for an estimated intake exceeds 20 rem, then extended or protracted treatment is strongly recommended, except for poorly transported material in an in the lung.

Decorporation therapy should be administered immediately following any suspected intake or accidental internal contamination in an in excess of established action levels. The extent and magnitude of an internal plutonium contamination usually cannot be determined quickly; however, the usefulness of therapy will diminish if plutonium is allowed to translocate to bone where DTPA is ineffective. La Bone (1994b) has provided a recent approach to evaluating urine data enhanced by chelation (DTPA) therapy.

An initial prophylactic chelation therapy may be appropriate because bioassay measurements (particularly urinalysis) cannot usually be completed within the response time required for effective chelation therapy. Urinalysis becomes very helpful following administration of chelation therapy because there is a direct correlation between DTPA, urinary excretion, and dose averted because of plutonium excreted. Bihl (1994) has shown that about 2 mrem of CEDE is averted for every dpm of  $^{239}\text{Pu}$  excreted. This provides a method of measuring the effectiveness of DTPA therapy and determining if it is worthwhile to continue therapy. For example, if DTPA is administered when untreated excretion is 2 dpm/d, excretion should increase to 20 to 100 dpm for a dose savings of 40- to 200-mrem/d CEDE. It is probable that the efficacy of treatment will decrease with continued administration as plutonium is removed from the liver and the rate of transfer into the systemic compartment decreases.

## **5.10 RESPONSE TO SUSPECTED INTAKES**

Experience has shown that most intakes of plutonium are accidental. Plutonium facilities and operating procedures are designed to prevent intakes. Nonetheless, it is important for management to prepare for the possibility that workers might receive an intake of plutonium--even though the probability of an incident may be very small. Prompt and appropriate action following an accidental intake of plutonium will allow for therapeutic measures to be taken to minimize the internal contamination and lessen the potential for harmful effects. The health physicist and medical staff should work closely to ensure that the proper course of action is followed.

All employees suspected of having received an intake of plutonium should be referred for special bioassay measurements. Because a fraction of an intake by inhalation may be retained in the nasal passages for a few hours after exposure to airborne radioactive materials, any level of contamination on a nasal swab indicates an intake that should be followed up by a special bioassay measurement program. However, lack of detection on nasal smears cannot be taken as evidence that an intake did not occur either because the nasal passages can be expected to clear very rapidly or, alternatively, because the worker could be a mouth-breather. Special bioassay

should also be initiated if plutonium contamination is found on the worker in an in the vicinity of nose or mouth.

For acute intakes, direct bioassay measurements should be taken before, during, and after the period of rapid clearance of activity. Urine and fecal samples collected after known or suspected inhalation incidents should also be used to estimate the magnitude of the intake. Initial assessment of intakes from contaminated wounds are based primarily on wound count and urinalysis data.

If a significant intake is indicated, the worker should not return to further potential exposure to plutonium until the intake has been thoroughly assessed and a predictable bioassay pattern established. This is particularly important because a new intake of a very low level may confound the interpretation of bioassay measurements for previous intakes of plutonium.

The health physicist must make important decisions for prompt action at the site of an accidental or suspected intake of plutonium or other radioactive materials. Often, these decisions must be based on limited data. Information that may be available for initially estimating the amount and type of intake may include the following:

- levels of measured contamination in an in the work area
- skin contamination levels, affected areas, and whether the skin is damaged or punctured
- wound contamination levels
- chemical form of the material involved
- results of air monitoring
- nasal smear activity levels
- sputum and/or mouth contamination.

The special bioassay monitoring program is initiated following a known or suspected intake. This information is needed for dose assessment and future exposure management. The intake is confirmed if follow-up bioassay measurements indicate positive measurement results. Additional bioassay measurements may be needed to quantify the intake and provide data for determining the effective dose equivalent. The frequency of bioassay monitoring will depend on the specific case to be evaluated. Selection of the appropriate sampling frequency is based on the previously discussed performance capabilities for workplace monitoring program, consultations with internal dosimetry specialists, and the cooperation of the affected employee.

#### **5.10.1 Planning**

The management at the plutonium facility should be prepared to follow an emergency action plan for response to a plutonium intake. If a worker accidentally inhales or ingests plutonium or is injured by a plutonium-contaminated object, the action plan should be initiated immediately. A rapid response is important because any delay in an implementing appropriate action could lessen the effectiveness of decorporation therapy and increase the probability for internalized plutonium to deposit on bone surfaces.

#### **5.10.2 Medical Response Plan**

The health physicist and medical staff must establish an emergency action plan for the appropriate management of an accidental intake of plutonium. The elements of the plan should include the following:

- Decision levels for determining when monitoring data or accident events require emergency medical response
  
- responsibilities of the affected worker, health physicist, medical staff, and management or supervisory personnel

- instructions for immediate medical care, decontamination, monitoring, and longer-term follow-up response
  
- provisions for periodically reviewing, updating, and rehearsing the emergency action plan.

The sequence and priority of the emergency action plan may vary with the magnitude and type of accidental conditions and their severity. An initial early assessment of the incident should focus, first, on treatment of life-threatening physical injuries and, second, on the radioactive contamination involved. Minor injuries should be treated after decontamination.

A rapid estimate of the amount of internal contamination by plutonium or other alpha-emitters may not be possible. If a significant intake (meaning one that exceeds 10 times the ALI) is suspected, medical staff should proceed with decorporation therapy after first treating major injuries.

### **5.10.3 Responsibilities for Management of Internal Contamination**

Responsibilities should be assigned for action in an in response to an accidental internal plutonium contamination. The affected worker has the responsibility to inform the health physicist, RCT, or his immediate supervisor as soon as an intake is suspected. (More broadly, all radiological workers have the responsibility to report conditions that could lead to an intake to their immediate supervisor and/or the health physics organization.) The health physicist or RCT should make an initial survey of the extent of the contamination and immediately contact his supervisor and, when action levels are exceeded, contact a member of the medical staff. He should continue to provide monitoring and radiation safety support to the medical staff and supervisors during the



management of the contamination incident. Care should be taken to limit the spread of radioactive contamination.

The health physicist should immediately begin to gather data on the time and extent of the incident. Contamination survey results should be recorded. Radionuclide identity, chemical form, and solubility classification should be determined. Nasal smears should be obtained immediately if an intake by inhalation is suspected. When action levels are exceeded, all urine and feces should be collected and labeled for analysis.

Decontamination should proceed with the assistance of the medical staff. Contaminated clothing and other objects should be saved for later analysis.

#### **5.10.4 Immediate Medical Care**

The medical staff should provide immediate emergency medical care for serious injuries to preserve the life and well-being of the affected worker. Minor injuries may await medical treatment until after an initial radiation survey is completed and the spread of contamination is controlled. However, the individual should be removed from the contaminated radiation area as soon as possible. Chemical contamination and acids should be washed immediately from the skin to prevent serious burns and reactions.

Use of a chelating agent should be considered immediately following an accidental intake of plutonium that exceeds the facility action levels (as suggested in an in Section 5.9). For maximum effectiveness, the chelating agent should be administered as soon as possible following the accidental intake of plutonium. Both the zinc or calcium salts of DTPA are approved for human use and are available under Investigational New Drug (IND) Permits for treating internal plutonium contamination. The worker to be treated must first be informed of the proposed use of an experimental drug, instructed on the purpose of administering the chelating agent, and warned about the possible side-effects of the drug. The worker must then give signed consent before DTPA chelation therapy may be initiated. Even though DTPA therapy is the only method available for reducing

the quantity of plutonium or americium retained in an in the body, the affected worker has the right to refuse its use.

The recommended therapy for decorporation is 1-g CA-DTPA or ZN-DTPA by intravenous injection or infused in an in 250-mL normal saline or 5% glucose in an in water, infused intravenously over 1 hour (Gerber and Thomas, 1992; NCRP, 1980). Treatment may be continued if bioassay indicates that decorporation therapy continues to enhance the urinary excretion of plutonium. Extended therapy has shown no ill effects (Carbaugh et al., 1989).

CA-DTPA should not be administered to potentially fertile female workers. Instead, ZN-DTPA should be used for internal decorporation of plutonium and other transuranic materials.

#### **5.10.5 Contaminated Wounds**

Medical treatment for contaminated wounds may include flushing with saline and decorporating solutions, debridement, and surgical excision of the wound. These measures are all the responsibility of trained medical staff operating under the direction of a physician. Health physics personnel can provide valuable assistance by prompt assessment of materials removed from the wound and identification of magnitude of residual activity as decontamination proceeds. Decontamination should continue until all radioactivity has been removed or until risk of permanent physical impairment is reached.

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## 6.0 EXTERNAL DOSE CONTROL

The purpose of an external dose control program is to protect the individual radiation worker by minimizing dose to levels as low as reasonably achievable (ALARA) and preventing exposures above prescribed limits. This also implies minimizing the collective dose by summing all the individual total effective dose equivalents in a specified population. This section discusses methods to minimize exposures by characterizing the radiations emitted by plutonium and effective methods to shield or otherwise reduce exposures.

The Department of Energy provides a detailed explanation of the recommendations for external dosimetry in the Implementation Guide. External Dosimetry Program (DOE, 1994b). Specific applicable documents for external dosimetry are listed in the reference list of that Implementation Guide. Because the requirements and recommendations are explicitly given in these documents, they will not be discussed in any great detail in this section. Rather, the emphasis will be given to items that are unique to plutonium facilities and the radiological aspects for safe handling of plutonium.

Measuring the external radiation exposure and the resultant dose for personnel handling plutonium is a difficult task because of the many radiations involved. Examples of the radioactive decay schemes and radiations emitted were presented in Section 2.0 for the various plutonium isotopes and radioactive progeny. Plutonium has a wide distribution of gamma energies; literally hundreds of different photon energies are present. Fortunately, plutonium emits few high-energy photons, so photon dose rates are low. But plutonium also emits highly penetrating neutrons from spontaneous fission and alpha-neutron reactions from compounds and alloys.

In the past, most of the dose in plutonium facilities was the result of plutonium production and fabrication operations. Most of these operations involved physical contact with freshly separated plutonium in glove boxes during fabrication and assembly operations. With the reduction in weapons production, emphasis has shifted to dismantlement and storage operations and to D&D of plutonium facilities. Much of the material in these facilities is low-exposure plutonium containing 6%  $^{240}\text{Pu}$  that is at least 20-years-old, so a significant fraction of the  $^{241}\text{Pu}$  has decayed

into  $^{241}\text{Am}$ . The radioactive progeny have increased gamma dose rates, making dismantlement of plutonium facilities more difficult. Although many of the examples in this section involve higher-exposure plutonium, it is expected that most dosage in plutonium facilities will originate from clean-up and storage of weapons-grade plutonium.

## 6.1 DOSE LIMITS

Limits of interest used for control of external radiations are specified at various depths by 10 CFR 835 (DOE, 1993c) as well as the ICRP and the National Council on Radiation Protection and Measurements (NCRP). The limits are given in Table 6.1 for the appropriate depths in tissue for the whole body, lens of the eye, “unlimited areas of skin,” forearms, and hands and feet.

**Table 6.1.** Effective Depth of Tissue for Various Organs

	Depth of tissue, mg/cm <sup>2</sup>
Deep (penetrating)	1000
Lens of eye	300
Shallow (skin)	7

### 6.1.1 Limiting Quantities

Recently, DOE has made significant changes in the methodology used for radiation protection. Previously, DOE used the concept of *dose equivalent*. For whole body irradiations, dose equivalent was the product of absorbed dose multiplied by the quality factor, which was evaluated by Monte Carlo calculations in a cylindrical phantom of 30-cm diameter and 60-cm height. For monoenergetic neutrons or photons normally incident on the phantom model, the dose equivalent was the highest value calculated anywhere in the phantom. Dose equivalent was non-additive because the maximum

values occur at different depths in the phantom for different energies. A detailed explanation of the calculations can be found in an article by Auxier et al. (1968).

In 1977, the ICRP introduced a major revision in recommended radiation protection with the introduction of ICRP Publication 26 (ICRP, 1977). The new methodology establishes a “risk-based” system of dose limitation. The ICRP introduced the terms *stochastic* and *nonstochastic* for radiation effects and set limits for both types of effect. Stochastic effects are defined as those for which the probability of the effect occurring (as opposed to the degree or severity of effect) is a function of radiation dose. Nonstochastic effects were defined as those for which the severity of the effect is a function of the dose; a threshold may exist. Limits were established such that the risk of stochastic effects occurring was equivalent to about the same risks faced by workers in “safe” industries who were not occupationally exposed to radiation in the workplace. Limits were also established for nonstochastic effects that prevented these effects from occurring even if the exposure occurred at the annual limit over the lifetime of the worker.

The ICRP specified in Publication 26 that radiation exposure be limited by the *effective dose equivalent*,  $H_E$ , which can be expressed by the relation:

$$H_E = \sum w_T D_T Q_T \quad (6.10)$$

where

$w_T$  = tissue weighing factor for the relevant organ or tissue T

$D_T$  = absorbed dose in the tissue or organ of interest

$Q_T$  = the quality factor averaged over the tissue or organ of interest.

The weighing factors are given in Table 6.2, which is taken from 10 CFR 835 (DOE, 1993c). Effective dose equivalent has the benefit that it is additive, and internal and external radiations can be added numerically to drive an overall estimate of risk.



**Table 6.2.** Tissue Weighing Factors

Tissue or Organ	Tissue Weighing Factor, $w_T$
Gonads	0.25
Breast	0.15
Bone marrow (red)	0.12
Lungs	0.12
Thyroid	0.03
Bone surfaces	0.03
Remainder <sup>(a)</sup>	0.30
Whole body <sup>(b)</sup>	1.00

(a) Remainder means the five other organs or tissues with the highest dose (e.g., liver, kidney, spleen, thymus, adrenal, pancreas, stomach, small intestine, and upper large intestine). The weighing factor for each remaining organ is 0.06.

(b) For the case of uniform external irradiation of the whole body, a weighing factor equal to 1 may be used in determining the effective dose equivalent.

The methodology of ICRP Publication 26 (ICRP, 1977) has been incorporated into 10 CFR 835 (DOE, 1993c), and into the Radiological Control Manual (DOE, 1994I). The annual radiation dose limits for DOE and its contractors are presented in Table 6.3.

However, DOE contractors usually establish lower annual administrative control levels, typically 500 mrem/year.



**Table 6.3.** Radiation Dose Limits for DOE and DOE Contractors

Type of Radiation Exposure	Annual Limit
<b>Occupational Exposures</b>	
Stochastic Effects	5-rem total effective dose equivalent from external sources and the CEDE intakes received during the year
Non-Stochastic Effects	
Lens of eye	15-rem dose equivalent
Extremity	50-rem dose equivalent
Skin	50-rem dose equivalent
Individual organ or tissue	50-rem dose equivalent
<b>Embryo/fetus of a Declared Pregnant Worker</b>	
Gestation period	0.5-rem dose equivalent
<b>Planned Special Exposure</b>	
Event plus Annual	5-rem total effective dose equivalent
Occupational exposure	
<b>Minors</b>	0.1-rem TEDE

In practice, it is very difficult to measure the effective dose equivalents specified in Table 6.3 because it is necessary to know not only the type of radiation but also its energy and direction. If the flux, energy, and direction of incidence are known, it is possible to calculate effective dose equivalent using fluence to effective dose equivalent conversion coefficients presented in ICRP Publication 51 (ICRP, 1987), which presents the effective dose equivalent as a function of energy for various irradiation geometries. Conversion coefficients for monodirectional beams of neutrons can be found in an article by Stewart et al. (1994). Conversion coefficients for photons in various irradiation geometries, including planar sources, can be found in a report by the Zankl et al. (1994). This will provide more accurate values of effective dose equivalent as opposed to numerically setting the value of effective dose equivalent equal to dose equivalent.

### 6.1.2 Operational Quantities

Because of the difficulties in determining effective dose equivalent from direct measurements, the concept of *operational quantities* has been introduced to be more closely related to measurable quantities. Operational quantities include *ambient dose equivalent* used for area monitoring and *personal dose equivalent* used for personnel dosimetry. Operational quantities are designed to be a conservative estimator of effective dose equivalent, i.e., the values of the operational quantities will be equal to or higher than the effective dose equivalent specified for the limiting quantities.

The ambient dose equivalent,  $H^*(d)$ , is the dose equivalent at a depth,  $d$ , in a 30-cm-diameter sphere of tissue, where a) the radiation field has the same fluence and energy distribution as the point of reference for the measurement and b) the fluence is unidirectional (i.e., the sphere can be viewed as being in an aligned radiation field). Most survey instruments are designed to measure ambient dose equivalent, and international standards are based on the ambient dose equivalent concept. The depth of interest is 1 cm of soft tissue, as specified in 10 CFR 835.2.

The personal dose equivalent,  $H_p(d)$ , is the dose equivalent in soft tissue at the appropriate depth,  $d$ , below a specified point on the body. Obviously, personnel dosimeters should be calibrated in terms of personal dose equivalent.

In reality, most instruments and personnel dosimeters used at DOE facilities are still calibrated in terms of dose equivalent. For example, consider the case in which personnel neutron dosimeters are calibrated on acrylic plastic phantoms at a specified distance from a calibrated neutron source. For DOELAP testing, the dose equivalent at this point has been calculated in accordance with NBS Special Publication 633, Procedures for Calibration of Neutron Personnel Dosimeters (Schwartz and Eisenhauer, 1982). These calculations are based on the Grundl-Eisenhauer energy spectrum and the conversion coefficients from NCRP Report 38 (NCRP, 1971), which are for the "old" values of dose equivalent from the cylindrical phantom calculations.

In most instances, the present methods based on dose equivalent over-estimate effective dose equivalent. In cases where personnel are approaching dose limits, it may be prudent to more accurately evaluate effective dose equivalent using special calibrations.

Depending on the irradiation geometry and energy, effective dose equivalent may be as much as a factor of two less than dose equivalent.

## **6.2 RADIATIONS IN PLUTONIUM FACILITIES**

As outlined in Section 2.0 of this report, plutonium emits a wide variety of radiations, including alpha and beta particles, as well as more penetrating X-rays and gamma rays. Because of the short half-life of  $^{241}\text{Pu}$ , the radioactive decay progeny are also important sources of radiation. This section outlines methods to calculate the dose equivalents from radiations emitted by plutonium and its progeny. Examples of measured dose rates are also included.

### **6.2.1 Alpha and Beta Doses**

Plutonium is primarily an alpha-emitter and is of great concern if inhaled, ingested or injected into the body. However, the skin is an effective barrier to alpha particles, and external contamination is only a problem if there is a wound or break in the skin.

Plutonium-241 is a beta-emitter that produces low-energy beta particles with a maximum energy of 0.022 MeV. Both alpha and beta particles are completely shielded by thin rubber gloves or other protective devices. The dose rate through a rubber glove originates primarily from the X-rays and low-energy photons generated from plutonium and  $^{241}\text{Am}$ , the decay progeny of  $^{241}\text{Pu}$ .

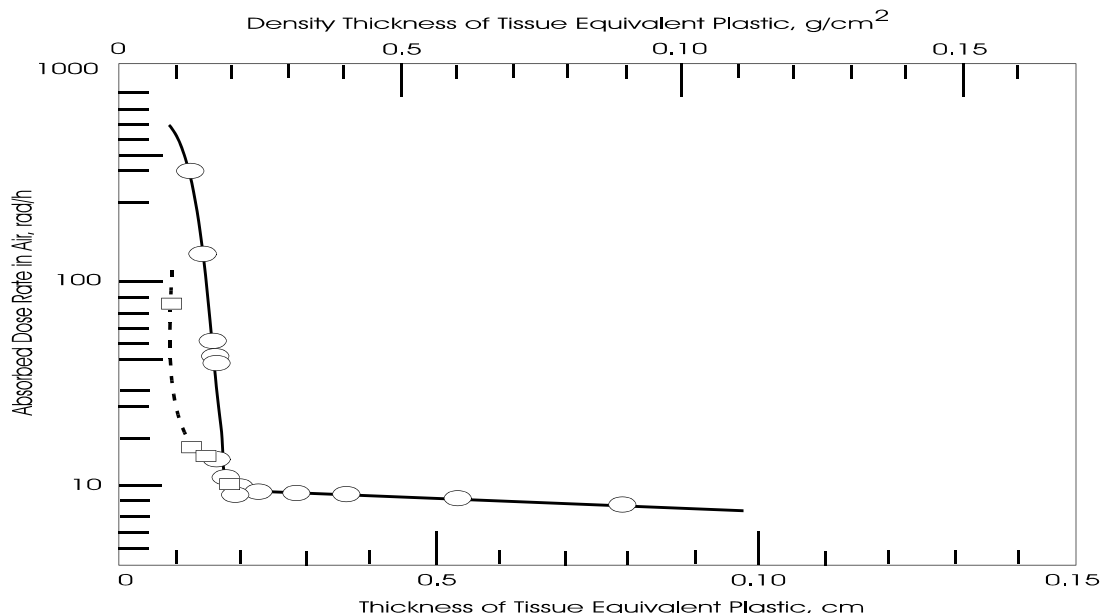
Figure 6.1 shows the dose rate as a function of tissue equivalent plastic absorber thickness, as measured by an extrapolation chamber in contact with a 3-in.-diameter plutonium dioxide source coated with a very thin layer of beryllium for contamination control. The plutonium was compressed to about 80% of its theoretical density and

vitrified by a Dynapack process in which powder was compressed into a glassy solid by extreme pressure and heat evolved during the compression process. The plutonium oxide disk is mechanically stable and produces little smearable contamination. Even minute layers of tissue equivalent plastic reduces the dose rate significantly, as shown in Figure 6.1.

### **6.2.2 Gamma Doses**

There can be substantial gamma doses involved in the processing and handling of plutonium, particularly in glove-box operations involving plutonium dioxide powders. Plutonium emits very few highly penetrating gamma rays; most of the radiations are L X-rays, which are very easily shielded. Because most of the photons emitted by plutonium are of low energy, plutonium sources are "infinitely thick" relative to their photon radiations, i.e., an additional thickness of plutonium does not appreciably increase the photon dose rate. A plutonium metal source of about 1-mm thickness or a plutonium oxide source about 6-mm-thick is "infinitely thick" due to self-shielding.

The age and isotopic composition are very important in determining the dose rate from plutonium because of the ingrowth of  $^{241}\text{Am}$  from the decay of  $^{241}\text{Pu}$ , which has a half-life of only 15 years. (The growth of plutonium daughters was discussed in detail in Section 2.1.1.) Old plutonium processing facilities can have high gamma dose rates, particularly from nearly invisible dust layers containing  $^{241}\text{Am}$ , which has a 37% probability of emitting a 60-keV photon per alpha disintegration. A surprising amount of plutonium oxide powder can be found in dust layers on the interior surfaces of glove boxes because of the very high density of plutonium. For example, a 0.001-in.-thick layer of plutonium oxide dust on the 4-ft by 8-ft floor of a glove box can contain almost 200 grams of plutonium. Even though a glove box has additional iron or lead shielding, high gamma dose rates can persist because of the photons emitted by dust layers on the surface of gloves. Covers must be placed over glove ports to reduce gamma dose rates around plutonium processing lines.



**Figure 6.1.** Absorbed Surface Dose Rate from Plutonium Dioxide as Measured with an Extrapolation Chamber

Doses to the extremities are usually dominated by gamma rays in typical glove-box operations. Extremity dosimeters must be used by all personnel who perform hand contact operations with plutonium or who are involved in the manual decommissioning of plutonium facilities. Extremities are defined as the hands and forearms below the elbows and the feet and legs below the knees. In a plutonium facility, the contact doses to the hands and forearms are the most limiting cases. The extremity dose is more limiting than a whole body dose if the dose gradient is greater than 10:1 over a distance of 1 meter, the maximum distance from the fingers to the trunk of the body. In most cases, the source is not at arm's length and the dose gradient needs to be 10:1 or 20:1 for the extremity dose to be limiting (NUREG/CR-4297, Reece et al., 1985). But in highly shielded glove boxes, it is possible to have very high extremity doses from dust layers on gloves; the dose to the torso can be much lower because of shielding applied to the glove box.

### **6.2.2.1 Measured Gamma Dose Rates**

There is a considerable amount of experimental data for measured photon dose rates from plutonium glove-box operations as recorded in progress reports issued by the Hanford Engineering Development Laboratory from the Personal Dosimetry and Shielding Program. For example, the photon dose rates were measured on an anthropomorphic Remab arm phantom inserted into gloves in a plutonium glove box. The arm phantom contains a human skeleton surrounded by tissue equivalent fluid inside a molded plastic "skin." Thermoluminescent dosimeters (TLD-700s) were positioned at various locations along the surface of the arm phantom and inside tubes inserted into the bones.

Measurements were first made in a "clean" glove box before it was placed into service. The arm phantom was placed inside the glove and positioned in contact with a 1-quart steel can (nominal wall thickness of 10 mil or 0.25 mm), containing 1 kg of plutonium dioxide with the isotopic composition shown in Table 6.4. Measurements were made at the various locations with the arm phantom inside 20-mil Neoprene gloves (average thickness 0.021 in., 0.53 mm) and inside 37-mil (0.94-mm) lead-loaded Neoprene gloves.

The data shown in Table 6.5 are the average dose rates measured by three TLD-700s with the indicated one standard deviation in the measured values. As one would expect, the palm and fingers had the highest dose rates, approximately 300 mrad/h; the lowest dose rates of 1 mrad/h were measured at the top of the arm. Because the plutonium was "infinitely thick" and lower-energy photons were removed by the shielding provided by the steel can, the dose rates in the lead-loaded glove were only slightly lower than those in the Neoprene glove. The can of plutonium was removed, and the gloves dusted with high-exposure plutonium with an isotopic composition similar to that given in Table 6.4. The arm phantom was inserted into 20-mil Neoprene and 37-mil lead-loaded Neoprene gloves; the dose rates measured with TLDs are shown in Table 6.6.



**Table 6.4.** Isotopic Composition of the Plutonium Used in the Extremity Dosimetry Measurements

Isotope	Weight Percent
$^{236}\text{Pu}$	0.000003
$^{238}\text{Pu}$	0.58
$^{239}\text{Pu}$	72.1
$^{240}\text{Pu}$	19.15
$^{241}\text{Pu}$	6.29
$^{242}\text{Pu}$	1.88
$^{241}\text{Am}$	0.02

**Table 6.5.** Gamma Dose Rates Along an Arm Phantom in Contact with a Steel Can Containing 1 kg of Plutonium Dioxide in an Uncontaminated Glove Box

Position	Gamma Dose Rates, mrad/h	
	Neoprene Glove	Lead-Loaded Glove
Ring finger	$330 \pm 6$	$272 \pm 25$
Palm	$292 \pm 9$	$220 \pm 16$
Back of hand	$72 \pm 2$	$65 \pm 1$
Wrist		
Inside	$84 \pm 6$	$56 \pm 5$
Outside	$31 \pm 1$	$24 \pm 1$
Forearm		
Inside	$22 \pm 0.4$	$12 \pm 1$
Outside	$4.4 \pm 0.1$	$3.8 \pm 0.4$
Elbow		
Inside	$4.8 \pm 0.1$	$2.6 \pm 0.2$
Outside	$1.4 \pm 0.1$	$1.8 \pm 0.4$
Front	$2.9 \pm 0.2$	$2.1 \pm 0.1$
Bottom of humerus	$2.2 \pm 0.1$	$2.5 \pm 0.5$
Lower mid-arm	$7.1 \pm 0.1$	$3.9 \pm 0.3$
Lower mid-humerus	$3.8 \pm 0.1$	$2.3 \pm 0.2$
Upper mid-arm	$2.4 \pm 0.1$	$2.5 \pm 0.2$
Upper mid-humerus	$1.8 \pm 0.1$	$1.8 \pm 0.2$
Top of arm	$0.9 \pm 0.03$	$2.2 \pm 0.8$
Top of humerus	$1.1 \pm 0.02$	$1.3 \pm 0.1$



As expected, the highest dose rates were recorded on the hand, wrist and forearm, where the most  $\text{PuO}_2$  dust had accumulated, and the lowest dose rates were on the upper arm and humerus. For thin dust layers, the dose rates inside the lead-loaded glove were generally much lower, typically a factor of 4 to 5 times less than the dose rates inside the Neoprene glove. The lead-loaded glove provided significantly better shielding for the 60-keV photons from  $^{241}\text{Am}$  and the L x-rays from plutonium, which were responsible for much of the dose. In these examples, the dose rates from the contaminated glove were about 10% of those from the 1 kg of plutonium dioxide inside the steel can. Additional experiments with 25%  $\text{PuO}_2$ -75% normal  $\text{UO}_2$  showed that dose rates increased as dust loadings increased with use; the dose rates on the hand and forearm increased to levels of about 30 mrem/h to 20 mrem/h, respectively.

The gamma energy spectra from plutonium sources are highly variable, depending on the amount of shielding present, including self-shielding. Small lightly shielded sources, such as dust layers on the interior of glove boxes, are dominated by L X-rays and the 60-keV photons from  $^{241}\text{Am}$ , the decay progeny of  $^{241}\text{Pu}$ .

**Table 6.6.** Gamma Dose Rates Measured with an Arm Phantom Placed Inside Gloves Dusted with Plutonium Dioxide Powder

Position	Gamma Dose Rates, mrad/h	
	Neoprene Glove	Lead-Loaded Glove
Palm	10.0 ± 0.4	9.5 ± 16
Back of hand	21.8 ± 1.3	5.4 ± 0.3
Wrist		
Inside	22.6 ± 0.7	9.0 ± 0.6
Outside	22.5 ± 0.6	5.8 ± 0.4
Forearm		
Inside	34.5 ± 0.2	6.7 ± 0.4
Outside	16.7 ± 0.2	3.6 ± 0.6
Elbow		
Inside	17.5 ± 0.4	5.3 ± 0.4
Outside	11.4 ± 0.1	3.7 ± 0.4
Front	---	4.4 ± 0.3
Bottom of humerus	3.5 ± 0.2	3.6 ± 0.4
Lower mid-arm	6.7 ± 0.5	3.4 ± 0.2
Lower mid-humerus	2.1 ± 0.2	2.9 ± 0.3
Upper mid-arm	4.6 ± 0.1	2.3 ± 0.4
Upper mid-humerus	1.0 ± 0.1	1.4 ± 0.3
Top of arm	0.8 ± 0.2	3.7 ± 0.3
Top of humerus	---	1.1 ± 0.3

But the gamma energy spectra are quite different in storage vaults and other facilities where the plutonium is encapsulated. In those cases, the low-energy photons have been shielded out, and the spectrum is dominated by higher photon energies. Note that plutonium metal buttons or cans of plutonium oxide prepared for storage are self-shielded, and high-energy photons from decay progeny such as  $^{237}\text{U}$  become increasingly important.

#### 6.2.2.2 Calculated Photon Dose Rates

It is very difficult to accurately calculate dose rates from plutonium because of the wide range of photon energies and the relatively low abundance of photons. Most of the photons are of relatively low energies, usually below 425 keV,

which are easily shielded. For heavily shielded spectra, the high-energy photons from decay progeny become very important, as well as the high-energy photons from plutonium, which have very low abundances.

For this reason, there are only a few computer codes that give accurate dose rates for plutonium. Many computer codes do not calculate the photons from progeny from radioactive decay. Others do not include the high-energy photons which have very low abundances, but which become very important for massive shields. One must check the photon libraries to make certain that the higher-energy photons are included. Also, many point kernel codes may not give accurate results for thin shields because low-energy build-up factors are not very accurate.

There are only a few codes specifically designed for plutonium dose calculations in the ORNL code center<sup>(a)</sup>; they include the following:

- PUSHLD - Calculation of Gamma Radiation Dose Rates from Three-Dimensional Plutonium Sources and Shield Geometries at Various Distances, HEDL-TME 73-89, Hanford Engineering Development Laboratory (Strode, 1974\_).
  
- BMC-MG - A Multigroup Monte Carlo Kernel Integration Neutron and Gamma-Ray Shielding Code System for Plutonium, BNWL-1855, Pacific Northwest Laboratory (Zimmerman, 1975\_).

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(a) RISC, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6362, Telephone (423) 574-6176.

- PURSE - A Plutonium Radiation Source Code, PNCT 852-78-13, Japan Power Reactor and Nuclear Fuel Development Corp., Tokai-Mura, Japan.

The PUSHLD computer code has the advantage that the calculated results were experimentally verified to make certain that the low-energy build-up factors were correct. There are undoubtedly several other codes that could give accurate dose rates from plutonium, particularly if a radioactive decay code is used to calculate the amount of progeny as a function of time. For example, the Los Alamos code MCNP (Briesmeier, 1986) would provide accurate neutron and photon doses if the decay progeny were included in the calculations. Unfortunately, all of these codes were developed years ago to operate on mainframe computers, and simple "user-friendly" versions are not available for personal computers.

There are some empirical equations that can be used to calculate dose rate through simple shields, such as Neoprene, when plutonium is directly handled in a glove box. Because of the dominance of low-energy X-rays, the surface dose rates from plutonium sources can be quite high. Roesch and Faust have derived a formula for predicting the surface dose rate from plutonium through a 100-mg/cm<sup>2</sup> shield:

$$D_s(\text{rad/h}) = 171 f_{238} + 0.51 f_{239} + 2.4 f_{240} + 8.7 f_{241} \quad (6.2)$$

$$+ 0.15 f_{242} (0.074 f_{241})t$$

where  $D_s$  = the surface dose rate of plutonium metal or oxide, rad/h

$f_i$  = the weight fraction of the  $i$ th isotope of plutonium

$t$  = the time since chemical separation of the plutonium, days.

This equation is only valid for a year or so after chemical separation, when the ingrowth of  $^{241}\text{Am}$  can be represented linearly.

A similar equation has been derived for lead-loaded rubber gloves using the calculations from the computer code PUSHLD. The 80-mil lead-loaded glove is nominally 1.9-mm (0.076-in.) thick in the palm and forearm and contains the equivalent of about 1 mm of lead. The surface dose rate,  $D_{\text{pbGl}}$ , is given by the following equation:

$$D_{\text{pbGl}}(t) = 2.83 f_{238} e^{-0.00789t} + 0.104 f_{239} + 0.0315 f_{240} \quad (6.3)$$

$$+ 6.35 \times 10^{-5} f_{242} + f_{241} (158.5 e^{-0.0016t} - 152.5 e^{-0.0457t})$$

where  $D_{\text{pbGl}}(t)$  = surface dose rate as a function of time, rad/h

$f_i$  = weight fraction of the  $i$ th plutonium isotope

$t$  = time since chemical separation of the plutonium, years.

This equation includes the radiations from plutonium, as well as the  $^{237}\text{U}$  and  $^{241}\text{Am}$  progeny from the decay of  $^{241}\text{Pu}$ . The expression is valid for times between 50 days and 100 years after the chemical separation of the plutonium. The formula predicts dose rates from 0% to +20% of those calculated by the computer code PUSHLD.

### 6.2.3 Neutron Dose Equivalents

Neutron dose equivalents are significant in any process or decommissioning efforts involving kilogram quantities of plutonium or gram quantities of  $^{238}\text{Pu}$ . Neutrons originate from three sources:

- Spontaneous fission of even isotopes of plutonium
- alpha-neutron reactions with low-atomic-number elements, including oxygen and fluorine in plutonium compounds and impurities in metals
- neutron-induced fissions.

Experience has shown that only spontaneous fission and alpha-neutron reactions are important. Because of strict criticality controls, most forms of plutonium have very little neutron-induced multiplication. Induced fission seems to be a problem only in metal (1 kg or more) or in very large, high-density arrays of plutonium oxide with an additional moderator.

Plutonium-238 used for heat sources deserves special attention. Even sub-gram quantities of  $^{238}\text{Pu}$  produce appreciable neutron doses because of the extremely high spontaneous fission rate in  $^{238}\text{Pu}$ . Also, the high specific heat of  $^{238}\text{Pu}$  creates handling problems; small microspheres of  $^{238}\text{Pu}$  can melt through gloves in glove boxes and produce contamination problems.

Plutonium compounds created during the plutonium manufacturing process can produce very high neutron dose rates, especially  $\text{PuF}_4$  created during the separation and purification of plutonium. Fluorinator glove boxes typically have the highest neutron

dose rates in a plutonium processing line. Although  $\text{PuO}_2$  is the preferred form because of its chemical stability, the oxide emits almost twice as many neutrons as pure metal. Neutrons are produced in alpha-neutron reactions with  $^{17}\text{O}$  and  $^{18}\text{O}$ . Some  $\text{PuO}_2$  sources used in medical applications are prepared with enriched  $^{16}\text{O}$  to reduce neutron dose rates, but isotopic enrichment is generally not used to reduce neutron doses from plutonium compounds.

### 6.2.3.1 Calculated Neutron Dose Equivalent Rates

Neutron dose equivalent rates can be calculated accurately with computer codes, such as MCNP (Briesmeier, 1986). The MCNP code has the advantage that it can calculate both neutron and photon doses through shielding and in complex arrays. The Monte Carlo codes can also calculate the effects of neutron multiplication in systems containing large amounts of plutonium.

However, neutron dose equivalent rates can also be calculated from simple empirical formulas. Unlike gamma doses, there is very little self-shielding for neutrons in subkilogram masses of plutonium. In most instances, a canister containing plutonium can be treated as a point source at the geometric center of the plutonium. The neutron dose equivalent rate from a plutonium source can be calculated by:

$$H = 0.0097 S/r^2 \quad (6.4)$$

where  $H$  = dose equivalent rate, mrem/h

$r$  = distance from the center of the source, cm

$S$  = neutron emission rate from the plutonium source.

The total neutron emission rate,  $S$ , is the product of the mass of plutonium (in grams) times  $Y$ , the total neutron yield per gram of plutonium (neutrons/second/gram) from spontaneous fission,  $(\alpha, n)$  reactions with low atomic number elements in contact with the plutonium, and fission-induced neutrons. Generally, fission-induced neutrons are required because stringent criticality safety rules prevent accumulation of enough moderator and plutonium to significantly increase neutron emission rates. But kilogram quantities of metals or compressed oxides can have significant multiplication and increased emission rates. For example, neutron emissions from 1 kg plutonium metal “buttons” are generally measured to be 30% higher than the calculated neutron emission rate.

#### **6.2.3.2 Neutron Emission Yields**

The neutrons produced by spontaneous fission and  $\alpha, n$  reactions can be estimated from the following information.

Most neutrons from spontaneous fission originate from the even plutonium isotopes:  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{242}\text{Pu}$ . Because it is the most abundant, the isotope  $^{240}\text{Pu}$  is the most important source of spontaneous fission neutrons. Decay progeny of plutonium have very low spontaneous neutron emissions. Table 6.7 contains spontaneous fission yields for plutonium and other isotopes that may be found in plutonium facilities within the DOE complex. These data are taken from NUREG/CR-5550 (Reilly et al., 1991) and are believed to be more current than the previously published PNL values (Faust et al. 1977, Brackenbush et al., 1988). As a rule of thumb, nuclides with even numbers of protons and neutrons have the highest spontaneous fission neutron emission rates. The spontaneous fission rate for odd-even nuclides is about 1000 times less, and the rate for odd-odd nuclides is about 100,000 less. Spontaneous fission neutrons are emitted with a Maxwellian energy distribution given by the equation:



$$N(E) \propto (\sqrt{E}) \exp(-E/1.43 \text{ MeV}) \quad (6.5)$$

where  $N(E)$  is the number of neutrons as a function of the energy  $E$  in MeV.

**Table 6.7.<sup>1</sup>** Spontaneous Fission Neutron Yields

Isotope	Total Half-Life	Spontaneous Fission Half-Life, years	Spontaneous Fission Yield, n/sec-gram
<sup>232</sup> Th	1.41 x 10 <sup>10</sup> y	>1 x 10 <sup>21</sup>	>6 x 10 <sup>-8</sup>
<sup>232</sup> U	71.7 y	8 x 10 <sup>13</sup>	1.3
<sup>233</sup> U	1.59 x 10 <sup>5</sup> y	1.2 x 10 <sup>17</sup>	8.6 x 10 <sup>-4</sup>
<sup>234</sup> U	2.45 x 10 <sup>5</sup> y	2.1 x 10 <sup>16</sup>	5.02 x 10 <sup>-3</sup>
<sup>235</sup> U	7.04 x 10 <sup>8</sup> y	3.5 x 10 <sup>17</sup>	2.99 x 10 <sup>-4</sup>
<sup>236</sup> U	2.34 x 10 <sup>7</sup> y	1.95 x 10 <sup>16</sup>	5.49 x 10 <sup>-3</sup>
<sup>238</sup> U	4.47 x 10 <sup>9</sup> y	8.20 x 10 <sup>15</sup>	1.36 x 10 <sup>-2</sup>
<sup>237</sup> Np	2.14 x 10 <sup>6</sup> y	1.0 x 10 <sup>18</sup>	1.14 x 10 <sup>-4</sup>
<sup>238</sup> Pu	87.74 y	4.77 x 10 <sup>10</sup>	2.59 x 10 <sup>3</sup>
<sup>239</sup> Pu	2.41 x 10 <sup>4</sup> y	5.48 x 10 <sup>15</sup>	2.18 x 10 <sup>-2</sup>
<sup>240</sup> Pu	6.56 x 10 <sup>3</sup> y	1.16 x 10 <sup>11</sup>	1.02 x 10 <sup>3</sup>
<sup>241</sup> Pu	14.35 y	2.5 x 10 <sup>15</sup>	5 x 10 <sup>-2</sup>
<sup>242</sup> Pu	3.76 x 10 <sup>5</sup> y	6.84 x 10 <sup>10</sup>	1.72 x 10 <sup>3</sup>
<sup>241</sup> Am	433.6 y	1.05 x 10 <sup>14</sup>	1.18
<sup>242</sup> Cm	163 days	6.56 x 10 <sup>6</sup>	2.10 x 10 <sup>7</sup>
<sup>244</sup> Cm	18.1 y	1.35 x 10 <sup>7</sup>	1.08 x 10 <sup>7</sup>
<sup>249</sup> Bk	320 days	1.90 x 10 <sup>9</sup>	1.0 x 10 <sup>5</sup>
<sup>252</sup> Cf	2.646 y	85.5	2.34 x 10 <sup>12</sup>

<sup>1</sup> Adapted from NUREG/CR-5550 (Reilly et al., 1991)

Energetic alpha particles can overcome coulomb barriers in low-atomic-number elements and create an unstable nucleus that emits neutrons. Because of the high alpha activity of plutonium, this can be a significant source of neutrons. There are two nuclear reactions that are of importance:



Table 6.8 contains the alpha-neutron yields for oxides and fluorides for the most common plutonium and transuranic nuclides. Note that the neutron yields are normalized per gram of nuclide, not per gram of compound. To obtain the yields per gram of compound, multiply by 0.88 for PuO<sub>2</sub> and 0.76 for PuF<sub>4</sub>. These data are taken from NUREG/CR-5550 (Reilly et al., 1991).

Table 6.9 contains the neutron yields for trace amounts of elemental impurities in plutonium metal or oxide. These data are also from NUREG/CR-5550 (Reilly et al., 1991) and are derived from thick target yields from accelerator data. The data in Table 6.9 differ from previous values in BNWL-2086 (Faust et al., 1977), and the authors have not experimentally checked the accuracy of these values. Two sets of data are included: one for alphas emitted from enriched uranium and the other for alphas emitted from <sup>239</sup>Pu. To determine the neutron yield from trace impurities, it is first necessary to determine the specific alpha activity from Table 6.8, and the neutron yield per parts per million per 10<sup>6</sup> alphas from Table 6.9 for either enriched uranium or plutonium. The specific neutron yield from impurities can be estimated from the following formula:

$$Y_{\text{imp}} = 10^{-12} A_{\alpha} \sum_j^n P_j I_j \quad (6.8)$$

where  $A_{\alpha}$  = alpha activity of the plutonium nuclides

$P_j$  = specific neutron yield from the jth element (neutrons/alpha-part per million) from Table 6.8

$I_j$  = elemental impurity concentration in plutonium (parts per million).

Note that this formula is valid only if the impurities are uniformly distributed with the plutonium so that the alpha particles directly interact with the impurities. Dust layers of plutonium oxide can also produce high neutron yields. For example, plutonium oxide dust layers on HEPA filters with borosilicate glass can produce neutron emission rates

10 times higher than those for pure oxide because of alpha-neutron reactions with boron in the glass fibers and aluminum spacer plates.

The total neutron yield per gram of plutonium can be found by summing the contributions from:

- Spontaneous fission (from Table 6.7)
- alpha-neutron reactions in oxides or fluorides (from Table 6.8)
- neutrons from low-atomic-number impurities (from Table 6.9).

Multiplying the specific neutron yield (neutrons/second-gram of plutonium) by the mass of plutonium (grams) gives S, the neutron emission rate (neutrons/second). The dose equivalent rate is then calculated using Equation 6.4.

**Table 6.8.** Neutron Yields from Alpha-Neutron Reactions for Oxides and Fluorides

Isotope	Alpha Decay Half-Life	Alpha Yield, $\alpha$ /s-g	Average Alpha Energy, MeV	$\alpha$ , n Yield in Oxides, n/s-g	$\alpha$ , n Yield in Fluorides, n/s-g
$^{232}\text{Th}$	$1.41 \times 10^{10}$ y	$4.1 \times 10^3$	4.00	$2.2 \times 10^{-5}$	
$^{232}\text{U}$	71.7 y	$8.0 \times 10^{11}$	5.30	$1.49 \times 10^4$	$2.6 \times 10^6$
$^{233}\text{U}$	$1.59 \times 10^5$ y	$3.5 \times 10^8$	4.82	4.8	$7.0 \times 10^2$
$^{234}\text{U}$	$2.45 \times 10^5$ y	$2.3 \times 10^8$	4.76	3.0	$5.8 \times 10^2$
$^{235}\text{U}$	$7.04 \times 10^8$ y	$7.9 \times 10^4$	4.40	$7.1 \times 10^{-4}$	0.08
$^{236}\text{U}$	$2.34 \times 10^7$ y	$2.3 \times 10^6$	4.48	$2.4 \times 10^{-2}$	2.9
$^{238}\text{U}$	$4.47 \times 10^9$ y	$1.2 \times 10^4$	4.19	$8.3 \times 10^{-5}$	0.028
$^{237}\text{Np}$	$2.14 \times 10^6$ y	$2.6 \times 10^7$	4.77	$3.4 \times 10^{-1}$	
$^{238}\text{Pu}$	87.74 y	$6.4 \times 10^{11}$	5.49	$1.34 \times 10^4$	$2.2 \times 10^6$
$^{239}\text{Pu}$	$2.41 \times 10^4$ y	$2.3 \times 10^9$	5.15	$3.81 \times 10^1$	$5.6 \times 10^3$
$^{240}\text{Pu}$	$6.56 \times 10^3$ y	$8.4 \times 10^9$	5.15	$1.41 \times 10^2$	$2.1 \times 10^4$
$^{241}\text{Pu}$	$5.90 \times 10^5$ y	$9.4 \times 10^7$	4.89	1.3	$1.7 \times 10^2$
$^{242}\text{Pu}$	$3.76 \times 10^5$ y	$1.4 \times 10^8$	4.90	2.0	$2.7 \times 10^2$
$^{241}\text{Am}$	433.6 y	$1.3 \times 10^{11}$	5.48	$2.69 \times 10^3$	
$^{242}\text{Cm}$	163 days	$1.2 \times 10^{14}$	6.10	$3.76 \times 10^6$	
$^{244}\text{Cm}$	18.1 y	$3.0 \times 10^{12}$	5.80	$7.73 \times 10^4$	
$^{249}\text{Bk}$	$6.6 \times 10^4$ y	$8.8 \times 10^8$	5.40	$1.8 \times 10^1$	
$^{252}\text{Cf}$	2.646 y	1.9 x	$10^{13}$	6.11	$6.0 \times 10^5$

**6.2.3.3 Quality Factors for Neutrons**

Approved Quality factors for neutrons are provided in 10 CFR 835.2. As used here Quality Factor means the principal modifying factor used to calculate the dose equivalent from the absorbed dose; the absorbed dose (expressed in rad or gray) is multiplied by the appropriate quality factor (Q).

The quality factors to be used for determining dose equivalent in rem for neutrons are as follows:

Neutron Energy	Quality Factor
$\leq 10$ keV	3
$> 10$ keV	10

When the spectral data are sufficient to identify the energy of the neutrons, the following mean quality factor values may be used:

## QUALITY FACTORS FOR NEUTRONS

[Mean quality factors, Q (maximum value in a 30-cm dosimetry phantom), and values of neutron flux density that deliver in 40 hours, a maximum dose equivalent of 100 mrem (0.001 sievert).]

Neutron energy (MeV)	Mean quality factor	Neutron flux density (cm <sup>-2</sup> s <sup>-1</sup> )
2.5 X 10 <sup>-8</sup> . . . . . Thermal	2	680
1 x 10 <sup>-7</sup> . . . . .	2	680
1 x 10 <sup>-6</sup> . . . . .	2	560
1 x 10 <sup>-5</sup> . . . . .	2	560
1 x 10 <sup>-4</sup> . . . . .	2	580
1 x 10 <sup>-3</sup> . . . . .	2	680
1 x 10 <sup>-2</sup> . . . . .	2.5	700
1 x 10 <sup>-1</sup> . . . . .	7.5	115
5 x 10 <sup>-1</sup> . . . . .	11	27
1 . . . . .	11	19
2.5 . . . . .	9	20
5 . . . . .	8	16
7 . . . . .	7	17
10 . . . . .	6.5	17
14 . . . . .	7.5	12
20 . . . . .	8	11
40 . . . . .	7	10
60 . . . . .	5.5	11
1 x 10 <sup>2</sup> . . . . .	4	14
2 x 10 <sup>2</sup> . . . . .	3.5	13
3 x 10 <sup>2</sup> . . . . .	3.5	11
4 x 10 <sup>2</sup> . . . . .	3.5	10

## **6.3 RADIATION DETECTION AND EVALUATION**

This section describes the response of portable instruments, personnel dosimeters, and nuclear accident dosimeters to the radiations emitted by plutonium, which are primarily photons and neutrons. Data are also included on special spectrometry instruments used to calibrate dosimeters in the field.

### **6.3.1 Response of Portable Survey Instruments**

The energy and angular responses of almost all portable gamma survey instruments have been well characterized and published in the instruction manuals available from the manufacturers. Because of the preponderance of low-energy photons, especially the 60-keV photons emitted by  $^{241}\text{Am}$ , particular attention should be given to the low-energy response.



**Table 6.9.** Neutron Yields for Trace Impurities in Plutonium and Uranium

Element	Neutron Yield	Neutron Yield	Average Neutron
	per 10 <sup>6</sup> Alphas	per 10 <sup>6</sup> Alphas	Energy in MeV
	at 4.7 MeV ( <sup>234</sup> U)	at 5.2 MeV ( <sup>239</sup> Pu)	for 5.2-MeV Alphas from Pu
Li	0.16 ± 0.04	1.13 ± 0.25	0.3
Be	44. ± 4	65. ± 5	4.2
B	12.4 ± 0.6	17.5 ± 0.4	2.9
C	0.051 ± 0.002	0.078 ± 0.004	4.4
O	0.040 ± 0.001	0.059 ± 0.002	1.9
F	3.1 ± 0.3	5.9 ± 0.6	1.2
Na	0.5 ± 0.5	1.1 ± 0.5	
Mg	0.42 ± 0.03	0.89 ± 0.02	2.7
Al	0.13 ± 0.01	0.41 ± 0.01	1.0
Si	0.028 ± 0.002	0.076 ± 0.003	1.2
Cl	0.01 ± 0.01	0.07 ± 0.04	

It is not generally well known that neutron survey instruments have a severe energy dependence. In fact, some manufacturers claim a  $\pm 15\%$  response per unit dose equivalent extending over an energy range of thermal to 15 MeV. The energy dependence of several commercially available neutron survey meters has been experimentally measured at the PTB in Germany (Liesecki and Cosack, 1984). Their measurements made with monoenergetic neutrons in low-scatter conditions demonstrate that a typical moderator-based neutron survey meter underestimates the dose equivalent by a factor of 2 at an energy of 14 MeV and overestimates dose equivalent by a factor of 2 to 3 at an energy of 20 keV. Survey instruments also exhibit changes in response with the direction of incidence of the neutrons due to absorption and scattering of the neutrons by the electronics package attached to the moderator/detector. This can also result in 40% variation in response, depending on the direction of incidence. Fortunately, plutonium compounds emit neutrons in the MeV range, where the problems with energy

and angular responses are minimal. Accuracies of  $\pm 15\%$  can be achieved with careful calibration with  $^{252}\text{Cf}$  or other fission sources.

### **6.3.2 Personnel Dosimetry**

The detailed requirements of an external dosimetry program are given in the Implementation Guide. External Dosimetry Program (DOE, 1994b). Explicit guidance and requirements are given and need not be repeated here. This section will focus on dosimetry problem areas specific to plutonium facilities and possible solutions.

Personnel working in plutonium facilities are exposed to both photon and neutron radiations, and plutonium processing is one of the largest contributors to neutron exposure in the United States. The response of beta-gamma personnel dosimeters is well documented and will not be discussed here.

The response of neutron dosimeters to the neutron fields encountered in the workplace must be evaluated. All existing neutron dosimeters have a severe energy response problem and must be carefully calibrated for the specific radiation field in which the neutron dosimeter is worn. Typically, neutron dosimeters are calibrated to bare  $^{252}\text{Cf}$  or  $\text{D}_2\text{O}$ -moderated  $^{252}\text{Cf}$  sources in a low-scatter calibration facility. Then, the neutron dosimeters are worn in plutonium facilities under high-neutron-scatter conditions, which produce a completely different energy spectrum than that in which the dosimeter was calibrated. Accordingly, a typical approach is to perform neutron energy field characterization surveys at selected areas in a facility. Based on these surveys a field correction factor is determined which corrects for the difference between the dosimeter response to the calibration source and the response to the neutron fields in the workplace. Because of the large response of TLD-albedo dosimeters to low-energy neutrons (with energies below about 20 keV), the response of the dosimeter usually depends on the scattering conditions rather than the initial neutron energy spectrum. These problems are discussed in detail in several documents, including PNL-3213, Personnel Neutron

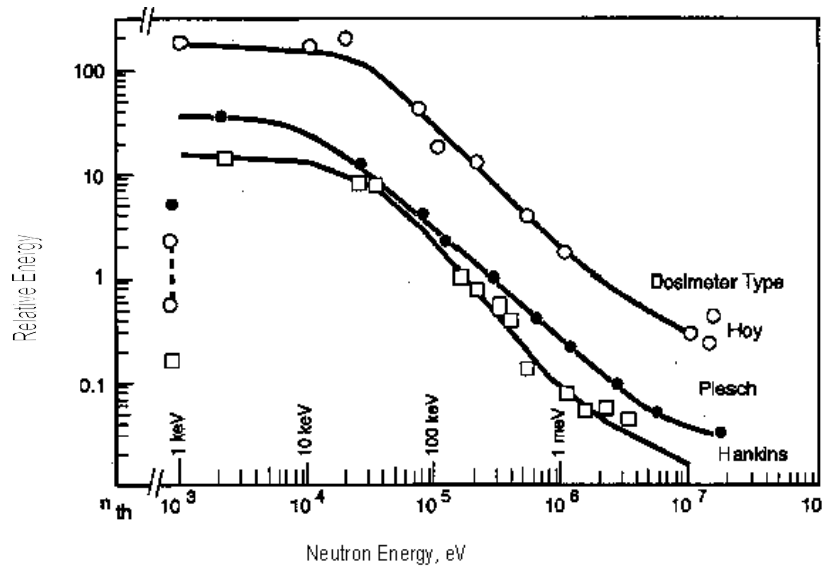
Dosimetry at Department of Energy Facilities (Brackenbush et al., 1980) and PNL-7881, Response of TLD-Albedo and Nuclear Track Dosimeters Exposed to Plutonium Sources (Brackenbush et al., 1991).

Thermoluminescent dosimeters are the most widely used neutron dosimeters in plutonium facilities. The energy response of a typical TLD-albedo dosimeter is shown in Figure 6.2. At neutron energies below about 20 keV, the energy response is almost constant. Above 20 keV, the response per unit dose equivalent drops dramatically by almost three orders of magnitude at 10 MeV. Almost all neutrons emitted by plutonium have energies in the MeV range. However, about 50% of the neutrons striking a thick concrete wall or floor are reflected back into the room at lower energies, and neutrons typically are reflected two or three times before being absorbed. Thus, the low-energy scattered neutrons are often more important in determining TLD-albedo dosimeter response than the high-energy neutrons emitted by the plutonium source. The TLD-albedo dosimeters are often calibrated in specific facilities by exposing them on phantoms at locations where the dose equivalent has been carefully determined from dose and spectrometric measurements (Brackenbush et al., 1991).

Recently, nuclear track dosimeters have been introduced for personnel dosimetry in plutonium facilities. These dosimeters have the advantage of a much more constant response per unit dose equivalent, as shown in Figure 6.3. Nuclear track dosimeters operate on the principle that a fast neutron interacts with plastic to produce a proton recoil that damages the polymer. Under special etch conditions, the damaged areas are removed to produce a distinct track, which is easily observed under a microscope. The neutron dose equivalent is then determined from the track density. Nuclear track dosimeters have a distinct threshold, usually about 100 keV.

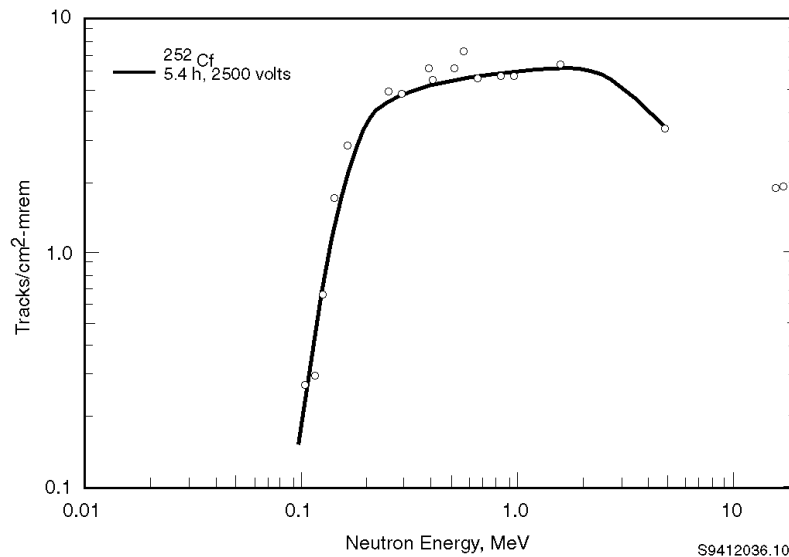
In conclusion, the combination of TLD-albedo and nuclear track dosimeters can provide a more uniform response with energy and more accurate personnel dosimetry. This combination of dosimeters may be an appropriate solution to neutron dose monitoring in DOE facilities with significant neutron exposure.

It is important to verify and document that personnel dosimetry systems provide accurate measurements and records of the occupational radiation doses received by workers in plutonium facilities (McDonald et al., 1992). To provide a level of confidence in dosimetry services in DOE facilities, the DOELAP accreditation program has been established. 10 CFR 835.402(b) (DOE 1993c) requires that personnel dosimetry programs shall be adequate to demonstrate compliance with 10 CFR 835.202, including routine dosimeter calibration and conformance with the requirements of the DOELAP for Personnel Dosimetry. The National Institute of Standards and Technology (NIST) has also established the National Voluntary Laboratory Accreditation Program (NVLAP) for testing and accreditation of dosimeter processors serving commercial industry and medical facilities. Because the dosimetry needs at many DOE facilities, particularly those processing plutonium, are different from commercial industries, the DOE established a more stringent accreditation program. The DOELAP standard includes some tests that are different from those in ANSI N13.11 (ANSI, 1983a) on which the NVLAP program is based. For example, the DOELAP standard has a test category for low-energy, nearly monoenergetic x-rays similar to those emitted by plutonium and americium. Both DOELAP and NVLAP accreditation programs use performance tests that evaluate the accuracy and precision of personnel dosimetry measurements. The accuracy is determined by comparing the measured dose equivalent to the “conventionally true dose equivalent” derived from calibration standards directly traceable to NIST in carefully controlled conditions.



**Figure 6.2.** Energy Dependence of Various TLD-Albedo Dosimeters

(Source: Piesch and Burgkhardt, 1978)



**Figure 6.3.** Response of Electrochemically Etched CR-39 Used in Nuclear Track Dosimeters as a Function of Neutron Energy

The general methods used to calibrate the dosimeters are given in the National Bureau of Standards Special Publication 633, Procedures for Calibrating Neutron Personnel Dosimeters (Schwartz and Eisenhauer, 1982). Two laboratories conduct the performance test irradiations for the DOELAP and NVLAP programs: Pacific Northwest Laboratory of Richland, Washington, and the Radiological and Environmental Laboratory (RESL) of Idaho Falls, Idaho. Dosimeters are submitted by processors for testing to the performance testing laboratories in the categories listed in Table 6.10. If the dosimeter processor passes certain accuracy and tolerance testing criteria, a team of dosimetry experts visit the processor and assess the operation of the dosimetry program, including dosimetry records and data retrieval systems, before the dosimeter processor is certified. These requirements are given in the U. S. Department of Energy Standard Laboratory Accreditation Program for Personnel Dosimetry Systems. DOE STD-1095-95 (DOE 1995h).

At present, only personnel dosimeters for whole body irradiations are tested, but a DOE working group has been formed to develop an extremity dosimetry performance testing standard. The DOE also conducts an intercomparison of calibration sources used for radiation protection purposes, but in the near future DOE secondary calibration laboratories will be established to increase the consistency of radiation protection instrument calibrations to national standards.

### **6.3.3 Extremity Dosimetry**

Doses to the extremities from plutonium processing and handling can involve significant exposures to the skin of the hands and forearms. Doses over small areas of the skin are discussed in Section IV.B.2. of Implementation Guide. External Dosimetry Program (DOE, 1994b) and will not be discussed here. That Section discusses skin contamination including hot particles, and the determination of skin dose from these events.

Highly accurate measurement of the dose to the hands and forearms is especially difficult because of the low-energy photons (L x-rays and 60-keV photons from  $^{241}\text{Am}$ ). Small variations in shielding, such as differences in the thickness of gloves used in glove boxes or nonuniform distribution of plutonium oxide dust on the surface of gloves, can produce large variations in the dose rate. Examples of these variations were given previously.

Extremity doses are typically determined by TLD finger rings, which are usually worn with the TLD chip facing the radiation source on the palm side of the hand. In glove-box and in D&D operations, the photon dose is usually significantly higher than any neutron dose. However, neutron dosimeters are sometimes worn to estimate extremity doses. Two types of neutron extremity dosimeters have been used: nuclear track dosimeters worn in special finger rings and specially calibrated TLD-albedo dosimeters worn on the wrist or forearm. DOE STD-1095-95 (DOE 1995h) is currently applicable to personnel dosimeters for whole body irradiation. However, it contains guidance for the development of correction factors for neutron doses that may be applicable for developing correction factors for neutron extremity dosimeters.

**Table 6.10.** Performance Test Categories, Radiation Sources, and Test Ranges for the DOELAP and NVLAP Programs

Category	Radiation Source	Test Range
Low-energy photons (high dose)	IST X-ray Beam code M150	0.1 - 50 Gy
High-energy photons (high dose)	<sup>137</sup> Cs	0.1 - 50 Gy
Low-energy photons (low dose)	NIST X-ray Beam codes: M30 M50 <sup>(a)</sup> S60 S75 <sup>(a)</sup> M100 <sup>(a)</sup> M150 H150 <sup>(b)</sup>	0.3 - 100 mSv
High-energy photons (low dose)	<sup>137</sup> Cs	0.3 - 100 mSv
Low-energy photons (monoenergetic)	15 - 20 keV <sup>(b)</sup> 55 - 65 keV <sup>(b)</sup>	0.3 - 50 mSv
Beta particles	<sup>204</sup> Tl <sup>90</sup> Sr/ <sup>90</sup> Y Natural or depleted uranium (slab) <sup>(b)</sup>	1.5 - 100 mSv 1.5 - 50 mSv
Neutrons	<sup>252</sup> Cf moderated <sup>252</sup> Cf unmoderated <sup>(b)</sup>	2.0 - 50 mSv
Photon mixtures		2.0 - 50 mSv
Photon/beta mixtures		
Photon/neutron mixtures		

(a) Category unique to the NVLAP program.

(b) Category unique to the DOELAP program. Note also that <sup>241</sup>Am (59-keV photons) may be used in place of the monoenergetic photon (55 - 65 keV) fluorescent X-ray source.

There is some question about the correct quality factor to apply to extremity neutron dosimeters. Existing quality factors are defined in terms of linear energy transfer (LET), so a numerical value for quality factor can be readily derived by calculation or measurement of the neutron energy spectra. However, the relationship between quality factor and LET was derived from biological experiments on cancer induction, especially



leukemia in blood-forming organs. There are no blood-forming organs in the extremities, so there is no biological basis for large values of quality factors for extremity exposures. However, regulatory agencies typically apply quality factors derived for whole-body exposures to the extremities, thus for compliance purposes quality factors should be applied for extremity exposures.

#### **6.3.4 Criticality Accident Dosimetry**

A criticality safety program, which includes material control, criticality alarms, and criticality accident dosimetry, is required as outlined in DOE Order 420.1 (DOE, 1996). The requirements in 10 CFR 835.1304 require that fixed nuclear accident dosimeters (NADs) and personnel nuclear accident dosimeters (PNADs) shall be worn by all personnel entering a controlled area that contains locations requiring an installed criticality alarm system, such as those required in DOE Order 420.1 (DOE, 1995c); which requires installed criticality alarms. The criticality accident dosimetry system should follow the provisions of ANSI/ANS 13.3-1981, Dosimetry for Criticality Accidents (ANSI, 1969a); this standard is currently being revised. Information on criticality accident dosimetry is also available from the International Atomic Energy Agency (IAEA, 1982).

The criticality accident program should contain the following items:

- a method and procedure to conduct an initial screening of personnel involved in a nuclear accident to determine whether significant exposures to radiation occurred (10 CFR 835.1304(b)(1))
  
- methods, procedures, and equipment for obtaining and analyzing biological materials (including  $^{24}\text{Na}$  activity from blood samples and  $^{32}\text{P}$  activity in the

hair)(10 CFR 835.1304(b)(2)), as well as metal coins, jewelry, and articles of apparel that may have become activated from neutrons

- a system of fixed dosimeters (i.e., NADs) (10 CFR 835.1304b(3)) capable of furnishing estimated radiation dose within an accuracy of  $\pm 25\%$  and the approximate neutron spectrum at the installed locations to allow conversion from rad to rem
- an operating range for the fixed dosimeters' neutron component 10 rad to 10,000 rad
- measurement capabilities for the fixed dosimeters' gamma ray component of fission gamma rays in the presence of neutrons with an accuracy of  $\pm 20\%$ , and an operating range for the gamma component operating range extending from 10 rad to 10,000 rad
- PNADs capable of furnishing sufficient information to determine neutron and gamma dose with an accuracy of  $\pm 25\%$  over a range of 10 rad to 1000 rad without dependence upon fixed NAD data
- a radiological counting laboratory with the methodology, analytical procedures, and quality assurance program in place to count the activated samples from the criticality accident and provide results quickly
- counting of activities in persons with significant exposures to assess the activation products in the body if a whole body counter is available (this is one of the more accurate methods for dose estimation)

- a health physicist designated to coordinate and evaluate the dosimetry information and provide dose estimates shortly after the accident
  
- a quality assurance program in place to help assure the accuracy and validity of the dosimetry results.

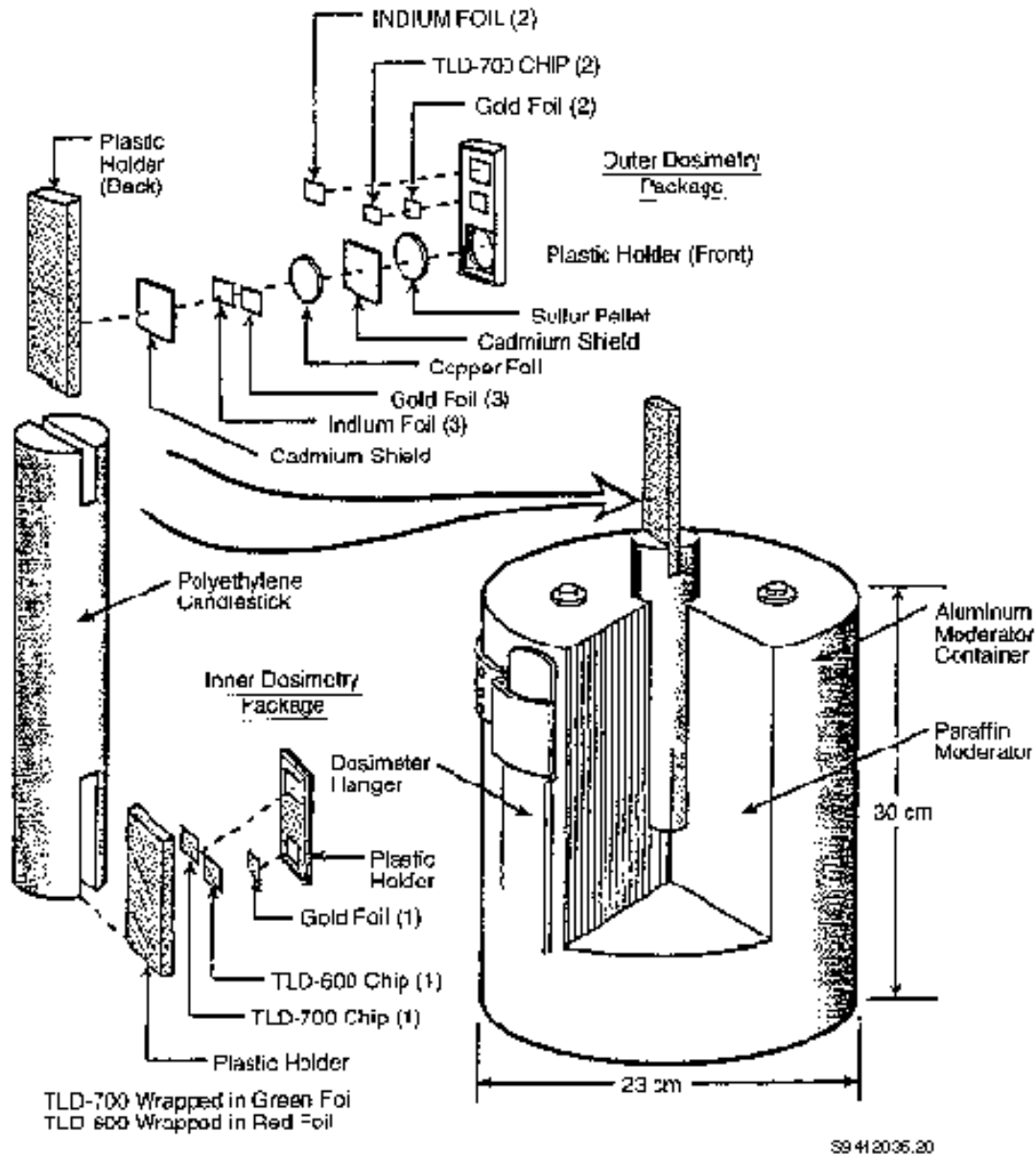
As mentioned in Section 6.1, the concept of dose equivalent was used to quantify exposures to different radiations. The quality factors used to determine dose equivalent are based on stochastic effects, primarily cancer induction some years later. But the doses in criticality accidents are typically so large that acute symptoms, including death, may occur within relatively short times, and quality factors are usually not applicable. For this reason, it is usually more appropriate to determine absorbed dose rather than dose equivalent if a person receives more than about 25 rem. These absorbed dose estimates to the torso are much more important for triage and treatment considerations.

The NADs are used to determine the neutron and photon dose at various locations in the plutonium facility, as well as providing spectral and calibration data for PNADs. A typical NAD used at the Hanford Site is shown in Figure 6.4. This unit is fixed to the wall or posted at locations around plutonium storage areas where it is easily recovered in the event of a criticality. The “candle” insert contains neutron- and gamma-sensitive TLDs as well as activation foils positioned at the center of the detector. Tests at the Health Physics Research Reactor at Oak Ridge have shown that this arrangement gives accurate estimates of “deep” dose for both neutrons and gamma rays. A set of foils identical to those used in the PNAD dosimeter is positioned above the moderator. These foils provide an estimate of the average cross-section or response per unit dose, so that the neutron dose from the foils in the PNAD can be more accurately evaluated for the incident neutron spectrum.

The PNAD dosimeter typically consists of several activation foils. In the case of the Los Alamos/Hanford design (Vasilik and Martin, 1981), the activation foils consist of ½-in.-

diameter foils of bare and cadmium-covered gold, bare and cadmium-covered indium, cadmium-covered copper, and a sulfur pellet. Algorithms have been developed to unfold an approximate neutron energy spectrum from the measured neutron activation products, so that neutron doses can be calculated. Criticality dosimeters containing various activation foils are available from vendors, but some of the commercial products do not contain sufficient material to measure neutron doses as low as 10 rad, which is the recommended lower detection limit for personal criticality accident dosimeters.

In the past, the Health Physics Research Reactor (HPRR) at Oak Ridge National Laboratory was available to calibrate criticality accident dosimeters, and several intercomparisons were held to demonstrate the accuracy and lower detection limit of criticality alarms and nuclear accident detector systems (Sims and Raga, 1987).



**Figure 6.4** Fixed Nuclear Accident Dosimeter Used at Hanford to help Assess Doses from Criticality Accidents

However, the HPRR has been dismantled. Some dosimeter testing capability is being established at pulsed reactor facilities at Sandia (Albuquerque, New Mexico) and at a Department of Defense pulsed reactor at the Aberdeen Proving Ground in Maryland.

### **6.3.5 Dose to Lens of Eye**

The dose to the lens of the eye is not generally a problem in plutonium facilities because whole body exposures are generally the limiting case. Dosimeters to measure the dose to the lens of the eye are seldom used. However, shallow doses at depths of 3 mm can be appreciable in cases where there is an abundance of low-energy photons, such as during visual inspection of machined plutonium pieces on laminar-flow tables or other situations where the plutonium is not shielded. In these cases, the eyes are generally protected by requiring safety glasses to be worn.

### **6.3.6 Spectrometry Measurements**

Personnel neutron dosimeters used at DOE plutonium facilities include TLD-albedo and nuclear track detectors. The response per unit dose equivalent for TLD-albedo dosimeters is a sensitive function of incident neutron energy (see Figure 6.2). These dosimeters are typically calibrated under low-scatter conditions in a calibration laboratory, such as the facility at PNL used for exposing dosimeters for DOELAP accreditation. The dosimeters are calibrated to a fission spectrum from  $^{252}\text{Cf}$  or a degraded fission spectrum from  $\text{D}_2\text{O}$ -moderated  $^{252}\text{Cf}$ . However, the neutron energy spectrum of the workplace is significantly different from that of the calibration facility and the response per unit dose equivalent is also different, primarily because of the number of low-energy neutrons produced by scatter within process equipment, glove boxes, and the walls and floor of the facility. To achieve accurate results, the TLD-albedo dosimeter results must be corrected for the specific neutron energy spectrum in which they are exposed. One method to achieve accuracy is to expose neutron

dosimeters on a phantom in the workplace in neutron fields where the dose equivalent rate has been carefully measured using neutron spectrometers.

There are several neutron energy spectrometers available to make accurate neutron spectrum measurements and dose equivalent estimations, as outlined in the document A Field Neutron Spectrometer for Health Physics Applications (Brackenbush et al., 1992). Neutron spectrometers that are useful for dose equivalent determinations in plutonium facilities include:

- Multisphere or Bonner sphere spectrometers
- tissue equivalent proportional counters (to determine LET spectra)
- liquid scintillator spectrometers
- proton recoil spectrometers.

#### **6.3.6.1 Multisphere Spectrometer System**

The multisphere or Bonner sphere spectrometer (Bramblett et al., 1960) is the neutron spectrometer system most often used by health physicists for neutron energy spectrum measurements, perhaps because it is simple to operate.

Multisphere spectrometers are typically used for measuring neutron energy spectra over a wide energy range from thermal energies to over 20 MeV although detailed energy spectra are not obtained. With the use of an appropriate spectrum unfolding code, the multisphere system will determine the average neutron

energy, dose equivalent rate, total flux, kerma, and graphical plots of differential flux versus energy and dose equivalent distribution versus energy.

The multisphere spectrometer consists of a set of polyethylene spheres of different diameters, typically 3 in. to 12 in. A thermal neutron detector, such as a  $^3\text{He}$  proportional counter or a  $^6\text{LiI}$  scintillator is positioned at the center of each sphere, and the count rate measured. The neutron energy spectrum can be determined from the ratio of counts from different detectors. However, the spectral unfolding algorithms do not provide mathematically unique solutions. The most appropriate solutions are obtained by making an initial guess that the spectrum consists of a fission spectrum with a  $1/E$  “tail.” Multisphere spectrometers have demonstrated accuracies of  $\pm 15\%$  when exposed to  $^{252}\text{Cf}$  sources with calibrations directly traceable to NIST (Brackenbush et al., 1991).

Figure 6.5 demonstrates the type of neutron energy spectra measured by the multisphere spectrometer. The plot shows the logarithmic plots of four multisphere spectrometer measurements made at a distance of 50 cm from 1 kg of plutonium for “bare” plutonium fluoride (i.e., no intervening shielding), plutonium fluoride shielded with 10 cm (4 in.) of acrylic plastic, “bare” plutonium oxide, and “bare” plutonium metal. The plutonium fluoride has the highest neutron emission rate and corresponds to the highest peak in the graph. The lowest peak corresponds to the moderated plutonium fluoride spectrum with 4 in. of acrylic plastic shielding. These measurements are typical of the neutron energy spectra in plutonium processing areas containing glove boxes. The spectra contain a significant fraction of low-energy scattered neutrons from the glove boxes and the thick concrete floor and walls of the facility. The spectra are distinctly different from neutron emission spectra (see Section 6.2), which do not contain scattered or background neutrons.



### 6.3.6.2 Tissue Equivalent Proportional Counter

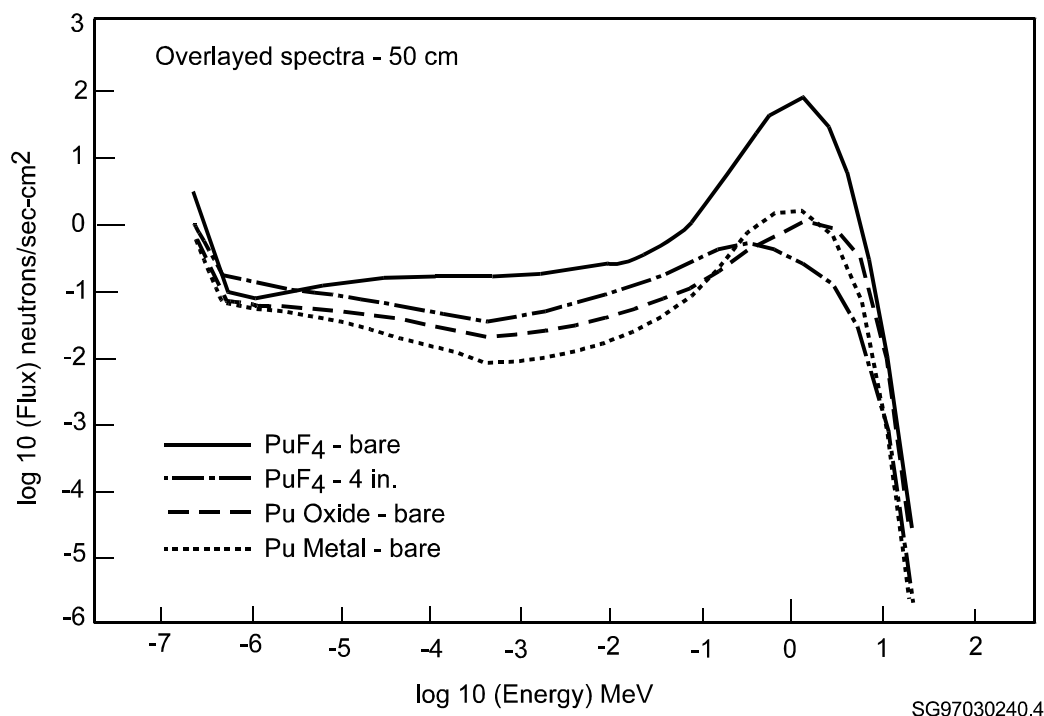
The tissue equivalent proportional counter (TEPC) is not often used by health physicists, but it can provide highly accurate estimates of dose equivalent. The TEPC consists of a hollow sphere or cylinder of tissue equivalent plastic filled with low-pressure equivalent gas. The pressure is so low (a few torr) that the TE gas cavity has the same mass stopping power as a 2- $\mu\text{m}$  sphere of tissue at unit density. Because the TEPC actually measures the energy absorption in a known mass of tissue equivalent material, it provides an absolute measure of absorbed neutron dose. The TEPC also measures the pattern of microscopic energy distributions from any penetrating ionizing radiation. With appropriate algorithms, LET distributions, hence quality factors, can be calculated. Thus, the TEPC provides absorbed dose, quality factor, and dose equivalent from a single spectral measurement of the event size distribution from the TEPC.

The TEPC can provide highly accurate measurements of dose equivalent under laboratory conditions. The TEPC can measure dose equivalent within  $\pm 5\%$  to  $\pm 10\%$  when exposed to NIST-calibrated  $^{252}\text{Cf}$  sources (Brackenbush et al., 1991). However, it suffers from stability problems, and its accuracy decreases with time as impurities diffuse from the TE plastic walls and temperature changes cause gain shifts in the proportional counter. Nevertheless, the TEPC can provide reasonably accurate measurements of dose equivalent in the workplace ( $\pm 15\%$ ) over extended time periods of 6 months or more, and can be used to monitor dosimeter irradiations on phantoms in the workplace.

### 6.3.6.3 Liquid Scintillator Spectrometer

The liquid scintillator spectrometer typically consists of a 2-in. by 2-in. cylindrical cell of hydrogenous scintillator solution in contact with a photomultiplier. Neutrons interact in the scintillator to produce proton recoils, which interact with the scintillator to produce light. With careful calibration, the incident neutron energy spectrum can be unfolded from the measured distribution of scintillation events.

The liquid scintillator spectrometer has the advantage that it is very sensitive and can operate at low dose rates. It is useful over an energy range extending from about 1 MeV to 20 MeV. Neutron dose equivalent can be calculated from the measured spectra using the conversion factors given in the Implementation Guide. External Dosimetry Program (DOE, 1994b). The dose equivalents calculated from liquid scintillator measurements are reasonably accurate ( $\pm 10\%$  to  $\pm 20\%$ ) for lightly moderated plutonium spectra. Because of the lower energy cut-off of liquid scintillator spectrometers, they may not provide accurate dose equivalent values outside heavily shielded facilities, such as plutonium storage vaults with thick concrete walls.



**Figure 6.5.** Neutron Energy Spectra as Measured by the Multisphere Spectrometer at 50 cm from Plutonium Metal, PuO<sub>2</sub>, and PuF<sub>4</sub> Sources

#### 6.3.6.4 Proton Recoil and Other Spectrometers

There are a number of other spectrometer systems that are available that have been used successfully for neutron spectral measurements in the field. Of particular interest is a spectrometer employing five different proton recoil proportional counters filled with gasses at different pressures. Each counter measures a different portion of the neutron spectrum, and quite accurate spectra can be obtained by “linking” all the measurements together. Unfortunately, this type of spectrometer is quite expensive, bulky, and difficult to use for workplace measurements.

Recently, neutron energy spectrometers have been fabricated from superheated drop detectors. Each type of superheated drop detector responds to neutrons with energies above a certain threshold. By using a combination of detectors with different thresholds and appropriate unfolding algorithms, it is possible to determine approximate neutron energy spectra from simple measurements. Commercially available units currently tend to suffer from quality control problems; i.e, the sensitivity per unit neutron fluence varies too much to make highly accurate spectral measurements. (The sensitivity varies with the degree of superheat and ambient temperature.) However, this technique offers great promise as a very simple, relatively inexpensive method for neutron spectrometry and dose determinations, particularly in facilities with a constant temperature.

## **6.4 EXTERNAL DOSE REDUCTION**

The traditional methods of using time, distance, and shielding are typically employed in plutonium facilities to reduce exposures to ALARA levels. However, other considerations may be just as important. Good housekeeping practices are vital to keep dose rates low. Even invisible dust layers on the interior surfaces of glove boxes can create gamma radiation fields of 10 mrem/h or more, especially through lightly shielded glove ports. The practice of pulling gloves outside for storage should not be condoned in operations that generate dust or powders. Dose rates of 30 mrem/h have been measured in facilities processing high-exposure oxide powders. A factor of 30 reduction in dose rate was achieved by merely storing the gloves inside the glove box when not in use and placing lightweight "pie plate" shields over the glove-port openings.

#### **6.4.1 Time**

Obviously, reducing the time a worker is exposed in a radiation field will reduce the dose. Any operation which involves high dose rates (more than a few mrem/hour) or long exposures should be reviewed for possible reductions in a worker's exposure time. For example, a worker should minimize the time spent near a fluorination operation. After the equipment has been set up, the worker should leave the area during the actual fluorination step.

#### **6.4.2 Distance**

Because of the inverse square relationship with discrete radiation sources, significant dose reductions can be achieved by increasing the distance between the worker and the plutonium source. Also, the low-energy photons emitted through glove ports and bag-out ports can be attenuated by several feet of air. Most plutonium operations involve contact work, so increasing the distance may not always be practical. But significant reductions in doses can be achieved by reducing plutonium inventories in glove boxes. It is good practice not to store plutonium samples in glove boxes, but to remove them to storage vaults or other shielded locations. In many cases, the plutonium samples can be stored in the glove box in "wells" or specially shielded areas at some distance from the work areas where the plutonium technicians spend most of their time. The best method of reducing neutron dose is simply to remove the plutonium from the glove box and minimize inventories in the glove box.

#### **6.4.3 Shielding**

The most practical method of reducing doses in plutonium operations is to apply shielding. Plutonium emits both neutrons and photons, which require different types of shielding materials to be effective. There are also additional constraints that must be met,

such as the maximum thickness of shielding that can be placed on glove boxes and still retain worker mobility. It has been found that more than about 8 cm (4 in.) of shielding on the exterior surface of a glove box greatly reduces the worker's manual dexterity and efficiency. It is also important to place the shielding close to the plutonium source and not to try to shield personnel. Because neutrons scatter around shadow shields, it is usually best to shield all surfaces of glove boxes or storage areas. The following sections describe the shielding effectiveness of common photon and neutron shielding materials.

#### **6.4.3.1 Photon Shielding**

Because of the preponderance of low-energy photons, significant reductions in gamma doses can be achieved by even modest shielding. It is important to note that there is a significant amount of self-shielding in plutonium samples. A 1-mm-thick plutonium metal sample is “infinitely thick” and additional thicknesses will not appreciably increase the dose rate. For this reason, the photon dose is more dependent on the surface area rather than on the mass of plutonium.

Invisible dust layers on gloves and interior surfaces of glove boxes can produce high exposure rates, especially if the gloves are pulled outside the glove box for storage to prevent them from being caught in machinery. Simple iron or lead shields placed over the glove ports can reduce the dose rates near the glove box by an order of magnitude. Modest gamma shields of 6 mm (0.25 in.) of lead and 13 mm (0.5 in.) of lead-loaded x-ray glass are usually sufficient to reduce photon dose rates from plutonium to acceptable levels.

Table 6.11 gives examples of how effective various gamma shielding materials are in reducing the dose rates from low-exposure (6%  $^{240}\text{Pu}$ ) and high-exposure (19%  $^{240}\text{Pu}$ ) sources. The sources consist of cylinders containing 1 kg of plutonium oxide; the dose rates are given at a distance of 2 m from the source. This example is typical of the shielding effectiveness for cans of plutonium containing kilogram quantities of plutonium oxide, as might be found in storage vaults.

In contrast, Figure 6.6 shows the reduction in photon dose rates from a small sample of plutonium oxide power weighing about 100 grams. The dose rates were measured at a distance of 3 cm from the surface of the plutonium, which was contained in polyvinyl chloride plastic bags (a total thickness of 33 mil or 0.85 mm) for radiation measurements. The isotopic composition of the plutonium was similar to that given in Table 6.12.

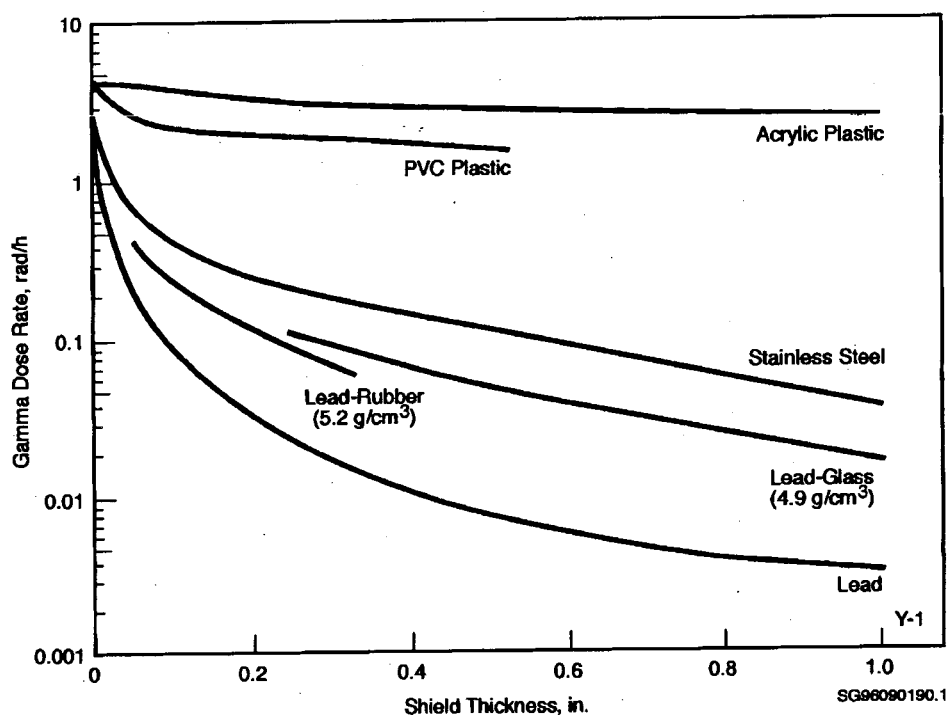
Photon radiation is a significant source of exposure, especially during D&D activities, when most of the plutonium has already been removed. Much of the photon exposure problem originates from thin dust layers, as described in the preceding paragraphs. High photon doses often originate from “streaming” through glove ports from dust layers on gloves. But there also can be appreciable neutron dose rates, even in supposedly “empty” glove boxes, from plutonium hold-up, especially in fluorinator glove boxes where there is a high neutron emission rate from alpha-neutron reactions. Wearing lead-loaded aprons can reduce dose rates by a factor of 2 in plutonium fuel manufacturing. High-exposure plutonium ( $>10\%$   $^{240}\text{Pu}$ ) should be handled in glove boxes with lead-loaded Neoprene gloves although some loss of mobility and dexterity may result. The photons from plutonium are easily shielded by several millimeters of lead or iron, but it requires almost 15 cm (6 in.) of polyethylene or hydrogenous moderator to reduce neutron doses by a factor of 10. Simplistically stated, the gamma dose rate is a function of surface area, while neutron dose rate is a function of the mass of the plutonium and its chemical form.

**Table 6.11.<sup>1</sup>** Photon Dose Rates at 2 Meters from Cylinders of Plutonium Containing 1 kg of Plutonium at 5 Years After Chemical Separation

Shield Material	Shield Thickness, cm	Photon Dose Rate, mrad/h	
		19% <sup>240</sup> Pu Source	6% <sup>240</sup> Pu Source
Polyvinyl chloride (PVC)	0.005	19.3	1.74
	0.038	8.72	0.570
	0.084	6.29	0.391
Lead glove	0.094	1.85	0.105
Heavy Lead glove	0.152	0.54	0.0464
Lucite	0.612	7.03	0.447
	2.54	3.30	0.190
	0.025	2.69	0.144
Steel	0.038	2.41	0.131
	0.051	2.19	0.121
	0.317	0.42	0.0418
	0.635	0.221	0.0299
	1.33	0.134	0.0205
	2.43	0.0766	0.0119
	0.635	0.0701	0.0103
	1.27	0.0380	0.00288
Lead	2.57	0.0156	0.000391
	5.08	0.00429	0.000023
	10.16	0.000467	0.0000001
	0.645	0.135	0.0251
	1.30	0.0841	0.0144
X-ray glass	2.60	0.0463	0.00534
	0.16	0.306	0.0346
	1.30	1.94	0.109
Safety glass	2.60	1.50	0.0886

<sup>1</sup> Adapted from NUREG/CR-5550 (Reilly et al., 1991)





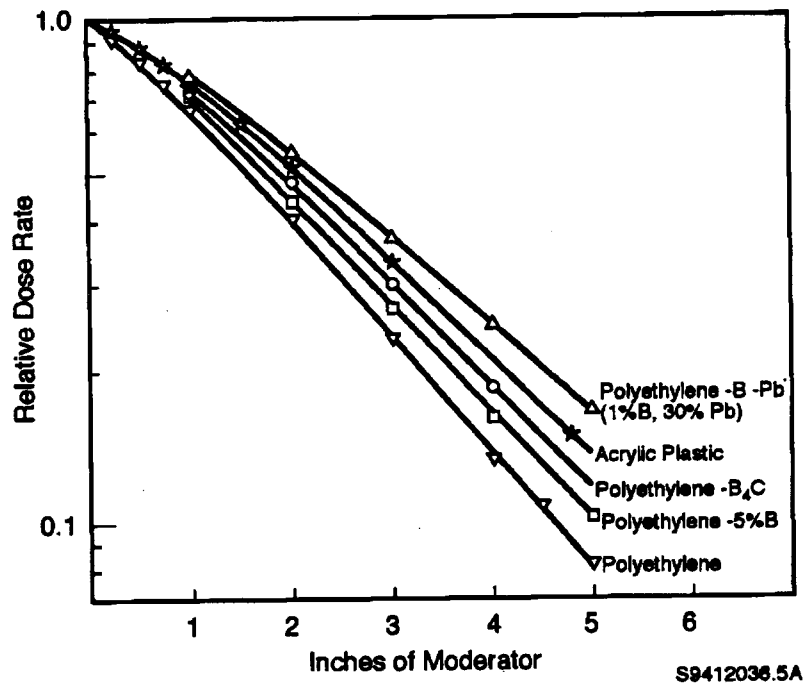
**Figure 6.6.** Reduction in Photon Dose Rate with Various Shielding Materials at a Distance of 3 cm from a 100-gram Disk of Plutonium Oxide

**Table 6.12.** Isotopic Composition of Plutonium Sources at 5 Years After Chemical Separation of the Plutonium

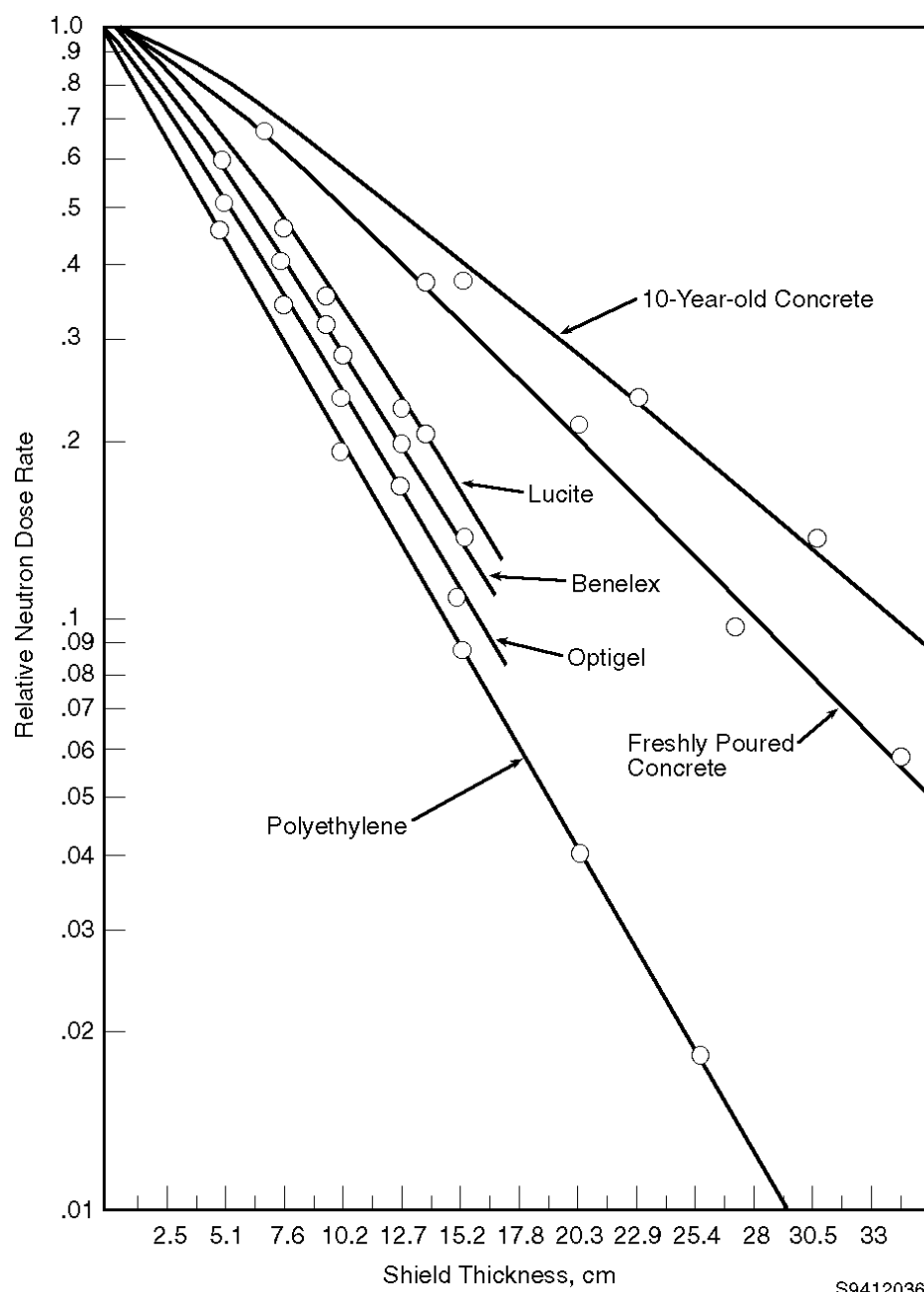
Isotope	Weight-Percent of Isotope	
	Low-exposure Pu	High-exposure Pu
<sup>238</sup> Pu	0.001	1.85
<sup>239</sup> Pu	93.5	63.3
<sup>240</sup> Pu	5.99	19.2
<sup>241</sup> Pu	0.397	9.27
<sup>242</sup> Pu	0.001	3.88
<sup>241</sup> Am	0.103	2.40

**6.4.3.2 Neutron Shielding**

The neutron radiations from plutonium are much more difficult to shield than the photon radiations. As a rule of thumb, it requires about 15 cm of hydrogenous shielding to reduce the neutron dose rate by an order of magnitude. Figure 6.7 shows the reduction in dose equivalent rate for various shielding materials for plutonium tetrafluoride sources, which have an average neutron energy of 1.3 MeV. For practical purposes, the shielding thickness for glove boxes is limited to about 4 in.; it is not possible to operate machinery through thicker shields. Figure 6.8 shows the reduction in dose equivalent rate through various slab shields for plutonium dioxide. These data were obtained from measurements of the neutron dose using a TEPC.



**Figure 6.7.** Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Tetrafluoride Sources



**Figure 6.8.** Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Oxide Sources

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## **7.0 NUCLEAR CRITICALITY SAFETY**

This chapter will emphasize present-day criticality concerns from the standpoint of what health physics personnel need to know to ensure that the DOE mission is accomplished in a safe and cost-effective manner. It provides an overview of the administrative and technical elements of current nuclear criticality safety programs. It does not provide a definitive discourse on nuclear criticality safety principles or repeat existing guidance. For health physics personnel who require a greater understanding of nuclear criticality safety, the references contained here provide a source of such detailed requirements and information.

Nuclear criticality safety issues at DOE facilities historically have been concerned with manufacturing plutonium, processing plutonium into weapon components, and storing weapon components and weapons in safe arrays. With DOE's newly identified mission of concluding much of the plutonium production and decommissioning of production reactors and processing facilities, today's nuclear criticality safety concerns have changed. While the historic nuclear criticality safety issues remain with the storage of weapons and associated components, current concerns include the disassembly of weapons, processing, and disposition of unique plutonium materials (commonly referred to as "legacy materials"), and decommissioning of production reactors and processing facilities.

Radiation protection personnel should understand nuclear criticality principles and the impact of these principles on radiological conditions that result from the processing, handling, and storage of fissionable materials. Radiation protection personnel provide an additional knowledgeable resource to help recognize workplace situations that might lead to the violation of a nuclear criticality control parameter that could contribute to an inadvertent nuclear criticality event. There have been occasions in the history of the nuclear industry when radiation protection personnel have observed and stopped unsafe actions by facility personnel that, if allowed to continue, might have resulted in an inadvertent nuclear criticality. Radiation protection personnel must also be aware of the potential impact of actions that may be routine for normal radiation protection practice, but which could result in the violation of a nuclear criticality control parameter. Finally, radiation protection personnel provide significant support in emergency response actions should an inadvertent nuclear criticality occur. These actions include use of emergency instrumentation, accident dosimetry, radiological dose assessment, and recovery.

This chapter reviews 1) nuclear criticality safety regulations and standards applicable to DOE facilities, 2) criticality control factors, 3) past criticality accidents and associated lessons learned, 4) roles, responsibilities, and authorities of health physics staff with regard to nuclear criticality safety, 5) the content of an acceptable nuclear criticality safety program, and 6) a summary of the criticality safety issues identified in DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

## **7.1 REGULATIONS AND STANDARDS**

Nuclear criticality safety program requirements for DOE facilities are presented in DOE O 420.1, Facility Safety (DOE, 1995c). The objectives for nuclear criticality safety in the Order are to establish nuclear criticality safety requirements. Specifically, these requirements shall (DOE Order 420.1, Section 4.3) ensure that:

1. Criticality safety is comprehensively addressed and receives an objective review, with all identified risks reduced to acceptably low levels and management authorization of the operation is documented.
2. The public, workers, property, both government and private, the environment, and essential operations are protected from the effects of a criticality accident.

DOE Order 420.1 incorporates the following American Nuclear Society ANSI/ANS Nuclear Criticality Safety Standards as requirements for DOE nuclear criticality safety programs. It may be noted however, that DOE Order 420.1 modifies certain sections of ANSI/ANS 8.1, 8.3, 8.5 and 8.7. This is a partial listing of incorporated standards that are of the most interest to radiation protection personnel:

- ANSI/ANS-8.1, Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors (ANSI, 1983b). This standard provides the basic criteria and limits for operations with fissionable materials outside reactors except for critical experiments. The standard also provides requirements for establishing the validity and the areas of applicability of any calculational method used in assessing nuclear criticality safety. It should be noted that the Order modifies certain paragraphs of the Standard.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- ANSI/ANS-8.3, Criticality Accident Alarm System (ANSI, 1986a). This standard provides the performance criteria for the location, selection, design, operation, and testing of nuclear criticality detection and alarm systems.
- ANSI/ANS-8.5, Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material (ANSI, 1986c). This standard describes the chemical and physical environment for the usage, properties of the rings and packed vessels, maintenance and test procedures, and criticality limits for solutions containing fissile materials.
- ANSI/ANS-8.7, Guide for Nuclear Criticality Safety in the Storage of Fissile Materials (ANSI, 1975a). This standard provides mass and spacing limits for the storage of, among other fissile materials, plutonium in both oxide and metallic forms.
- ANSI/ANS-8.15, Nuclear Criticality Control of Special Actinide Elements (ANSI, 1981). This standard provides the single nuclear criticality control parameter limits for the unique aspects of the special actinides (e.g., certain neptunium, plutonium, americium, and curium isotopes).
- ANSI/ANS-8.19, Administrative Practices for Nuclear Criticality Safety (ANSI, 1984). This standard provides the elements of an acceptable nuclear criticality safety program for operations outside of reactors.

Additional Standards of interest that are not required by DOE Order 420.1 include:

- ANSI/ANS-8.20, Nuclear Criticality Safety Training (ANSI, 1991). This standard (referenced in DOE Order 5480.20A) provides the criteria for the administration of a nuclear criticality safety training program for personnel who manage, work in or near facilities, or work outside of reactors, where the potential exists for nuclear criticality accidents.
- ANSI/ANS-10.3, Documentation of Computer Software (ANSI, 1986b). This standard presents guidelines for documenting computer codes (i.e., user documentation) for engineering and scientific applications.
- ANSI/ANS-10.4, Guidelines for the Verification and Validation of Scientific and Engineering Computer Programs for the Nuclear Industry (ANSI, 1987a). The objective of this standard is to

identify processes that will enhance the reliability of computer codes used in the nuclear industry and reduce the risk of incorrect application.

## **7.2 CRITICALITY CONTROL FACTORS**

For a criticality accident to occur, there must be a critical mass of fissionable material. As noted in ANSI/ANS-8.1 (ANSI, 1983b), the critical mass is a function of the radionuclides in the material as well as its density, chemical and physical form, shape, and surroundings (i.e., moderators, reflectors, neutron absorbers). Nuclear criticality safety is achieved through the control over both the quantity and distribution of fissile materials and other materials capable of sustaining a chain reaction as well as the control of the quantities, distributions, and nuclear properties of all other materials with which fissile materials are associated. For new facilities, DOE requires that design considerations for the establishing controls should be mass, density, geometry, moderation, reflection, interaction, material types, and nuclear poisons (neutron absorbers). The use of administrative controls is to be minimized (DOE, 1995c).

Nuclear criticality control factors can be classified as technical (e.g., geometry controls and mass-limitation controls) or administrative (e.g., operating procedures).

### **7.2.1 Technical Control Factors**

Plutonium isotopes include  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$ . All these radionuclides are fissionable materials; however,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  are referred to as fissile materials, a subset of fissionable materials. Fissile materials are capable of sustaining a neutron chain reaction with thermal neutrons and fast neutrons and, as such, have lower critical masses than other plutonium isotopes.

Single-parameter limits for plutonium solutions, oxides, and metals are presented in ANSI/ANS-8.1 (ANSI, 1983b) and ANSI/ANS-8.15 (ANSI, 1981) and are summarized in Table 7.1. A single-parameter limit means that if any one of the parameters for a given material is maintained less than its limit, then a criticality event is impossible. For example, for a  $^{239}\text{Pu}(\text{NO}_3)_4$  solution, as long as the  $^{239}\text{Pu}$  mass in the solution is less than 0.48 kg, the other parameters can exceed their limits (e.g., the solution concentration

could be greater than 7.3 g/L) and a criticality incident is not possible. The reader is referred to ANSI/ANS-8.1 (ANSI, 1983b) for a discussion of multiparameter control.

For plutonium solutions and metals in an isolated system, use of favorable geometry is the preferred method of criticality control. An isolated system is far enough removed from other systems such that neutron leakage from a nearby system will not contribute to the likelihood of a criticality excursion. Where geometry control is not feasible, the preferred order of controls is 1) other passive engineering controls (e.g., mass control), 2) active engineering controls, and 3) administrative controls. DOE Order 420.1 requires that the basis for not selecting geometry control be documented.

Other technical control factors used to control nuclear criticality risks include density controls, spacing controls (sometimes referred to as interaction), neutron absorbers, moderation controls, and neutron reflection. Spacing controls become particularly important in the storage and transport of fissionable materials. ANSI/ANS-8.1 provides additional discussion of technical control factors.

### **7.2.2 Double Contingency**

DOE Order 420.1 addresses the concept of the application of double contingency in nuclear criticality safety. This principle applies the technical control parameters above to ensure nuclear criticality safety. This is referred to as double contingency and must be adhered to at DOE facilities.

Double contingency requires that process designs incorporate sufficient factors of safety to require at least two unlikely, independent, and concurrent changes in process conditions before a nuclear criticality accident is possible.



**Table 7.1.** Subcritical, Single Parameter Limits for Plutonium Solutions and Metals (ANSI, 1983b)

Parameter	Plutonium Solutions and Metals			
	$^{239}\text{Pu}(\text{NO}_3)_4$	Metallic $^{239}\text{Pu}$	$^{239}\text{PuO}_2^{(a)}$	$^{239}\text{PuO}_2^{(b)}$
Mass of fissionable nuclide, kg	0.48	5.0	10.2	27
Diameter of cylinder of solution, cm	15.4	-	-	-
Volume of solution, L	7.3	-	-	-
Concentration of fissionable nuclide, g/L	7.3	-	-	-
Cylinder diameter, cm	-	4.4	7.2	12.6
Slab thickness, cm	5.5	0.65	1.4	2.8
Maximum density for which mass and dimension limits are valid, g/cm <sup>3</sup>	-	19.82	9.92	-

(a) Oxides containing no more than 1.5% water by weight at full density.

(b) Oxides containing no more than 1.5% water by weight at no more than half density.

Protection, or defense in depth, should be provided by either (a) the control of two independent process parameters (which is the preferred approach, if practical) or (b) a system of multiple controls on a single parameter. In all cases, no single credible event or failure shall (DOE Order 420.1, Section 4.3.3) result in the potential for a criticality accident. The basis for selecting either approach are to be fully documented.

The two parameters that are controlled in the double contingency analysis process must not be related by common mode failures. Judgment is required in determining whether two events are related and, consequently, whether they represent two contingencies or a single contingency. For example, exceeding a storage limit and then flooding an area with water would constitute two independent events. However, a fire followed by the flooding of a storage area with fire suppression water would constitute a single event.

The double contingency principle is to be applied to all nuclear criticality safety analyses for processes, systems and equipment, storage, and transportation of fissile materials.

Should contingencies be determined to be related, efforts are to be made to separate the contingencies. If this is not practical, an exemption process is identified in DOE Order 420.1.

### **7.2.3 Administrative Control Factors**

Administrative control factors are the combination of personnel, programs, plans, procedures, training, audits and reviews, and quality assurance practices which are used to administer a nuclear criticality safety program. Administrative controls are used in addition to engineered controls or design features to ensure nuclear criticality safety of facility operations. Administrative control factors are outlined in ANSI/ANS-8.19 (ANSI, 1984). An effective nuclear criticality safety program requires a joint effort by managers, supervisors, plutonium workers, and nuclear criticality safety staff and relies upon conformance with operating procedures by all involved personnel. The following sections describe the key requirements of a nuclear criticality safety program from ANSI/ANS-8.19.

#### **7.2.3.1 Nuclear Criticality Safety Program**

Management shall develop a nuclear criticality safety policy and ensure that it is distributed to fissile material workers. They also delegate authority to implement the policy, monitor the nuclear criticality safety program, and periodically participate in audits of the program. Supervisory staff shall ensure that nuclear criticality safety procedures are written and that staff is trained in those procedures. The nuclear criticality safety staff shall provide technical guidance for the design of equipment and processes and for the development of operating procedures. A nuclear criticality safety evaluation shall be performed by the nuclear criticality safety staff before starting a new operation with fissile materials or before an existing operation is changed. An independent evaluation of the technical adequacy of the nuclear criticality safety program shall also be performed periodically.

**7.2.3.2 Nuclear Criticality Safety Organization**

Like the radiation protection program, the nuclear criticality safety organization should have a reporting line to the highest level of facility management independent of operations. The nuclear criticality safety organization shall have the responsibilities and authorities of its staff clearly delineated and communicated to the other facility personnel. Lines of interaction and interfaces with other facility organizational components should be clearly defined, both organizationally and procedurally. In any case, the responsibility for nuclear criticality safety should be assigned in a manner that is compatible and consistent with the other safety disciplines. The organization should also contain an independent nuclear criticality safety review committee and have access to consultants to assist in the conduct of the criticality safety program.

**7.2.3.3 Plans and Procedures**

Facility nuclear criticality safety plans and procedures are important components of the overall facility operation. These documents provide the means by which the program is conducted and prescribe how nuclear criticality safety is to be achieved. These plans and procedures identify how both the administrative activities are to occur and how the technical aspects of nuclear criticality safety analysis are conducted. The purpose of procedures is to facilitate the safe and efficient conduct of operations. The processes of procedure development, review, training, and approval have sufficient controls to ensure that nuclear criticality concerns are properly addressed. These controls include the periodic review and reaffirmation of these procedures, ensuring that procedure deviations are properly investigated and reported to facility management and, if appropriate, to DOE. The controls should also mitigate the possibility of such deviations recurring.

Procedures should exist that address the determination and posting of nuclear criticality safety parameters. These procedures should include a description of how the limits are to be determined and how workstations are to be posted as to form, geometry controls, mass limits, moderator limits, etc.

Inspections and audits are performed to assess the success of the nuclear criticality safety program. The audits must be performed by qualified individuals who are independent of the operation. They are conducted to verify that operating procedures and other safety standards are being followed and to identify any weaknesses in the nuclear safety program. Deficiencies identified in these inspections and audits must be formally addressed, tracked, reported, and resolved.

ANSI/ANS-8.20 (ANSI, 1991) provides guidance for development of nuclear criticality safety training plans and procedures for personnel working with or near fissile materials. This program and its associated procedures should describe the program, training requirements, recordkeeping, content, responsibilities, and objectives of a facility nuclear criticality safety program.

#### **7.2.3.4 Nuclear Facility Safety Analysis**

Safety analysis reports document the analysis and the potential consequences of accidents and abnormal occurrences at nuclear facilities. For those facilities which process, store, and handle plutonium and other fissile materials, nuclear criticality safety analysis is a required element of the facility safety analysis report. The process includes the identification of hazards in the facility (including nuclear criticality safety hazards), the identification and development of potential scenarios involving nuclear criticality concerns, the development of failure modes and the potential effects of the accident, and the consequences of the accident. This safety analysis report, and the associated technical safety requirements, should document both the entire nuclear criticality safety program and the analysis process to assure the reviewer that nuclear criticality safety concerns are being properly addressed at the facility.

### 7.3 CRITICALITY ACCIDENT EXPERIENCE

Criticality accidents, sometimes called criticality excursions, can either be short-duration pulse-type excursions or continuous excursions. In the history of plutonium handling and processing, there have been five criticality accidents involving plutonium materials. Three of the accidents occurred during research activities and the other two accidents during plutonium-processing operations. The two processing accidents are reviewed in this section.

#### 7.3.1 Types of Criticality Accidents

In a pulse-type criticality accident, there is an initial pulse of typically  $10^{15}$ - $10^{17}$  fissions over a short time-period (less than 1 sec), sometimes following by additional lower-intensity pulses. In a fissionable material solution, the pulse or spike is terminated by the heating and consequent thermal expansion of the solution and by bubble formation that serves to reconfigure the fissile mass into a noncritical configuration (Paxton, 1966). If the initial pulse results in a loss of solution from the container (e.g., by splashing) or redistribution of material, the criticality event may conclude without further pulses. However, if there is no loss of material as the solution cools, it may form a criticality mass once again and pulse with slightly lower fission yield (Paxton, 1966).

Criticality accidents can result in lethal doses of neutron and gamma radiation at considerable distances from the accident site (on the order of tens of meters). There can also be high beta-gamma residual radiation levels from fission products after the excursion is concluded. The heat generated during the excursion can melt parts of the system that held the fissionable material (Moe, 1988).

Moe (1988) reviewed estimated prompt radiation doses from excursions in a moderated system and a metallic system, as well as dose rates from residual contamination left by a criticality excursion. Assuming a burst of  $10^{18}$  fissions in an unshielded, water-moderated system, the total absorbed dose is estimated to be >600 rad up to 6 m and >100 rad up to about 15 m. The gamma/neutron ratio of the total absorbed dose was 2.8. The gamma/neutron absorbed dose ratio was 0.1. In general, for a moderated system, the gamma dose would be expected to be higher than the neutron dose and, for a metal system, the neutron dose would be expected to be higher than the gamma dose.

Moe (1988) noted that for an excursion of  $>10^{18}$  fissions, dispersion of the fissile material and the fission products would occur, resulting in heavy local contamination and a subsequent high residual dose rate. This dose rate was estimated at  $>1000$  rad/h at 100 ft shortly after the burst and  $>10$  rad/h at 30 ft an hour after the burst. This is the basis for instructing workers to immediately run from the work area when the criticality alarm is sounded. Seconds can save significant dose, if not from the excursion itself, then from any residual radiation that is in the area.

### **7.3.2 Summary of Past Criticality Accidents**

Historically, there have been a total of five criticality accidents involving plutonium (Stratton, 1967). Three of the accidents involved plutonium in solutions, with the other two involving metallic forms. Three of the accidents involved early research activities and the other two were plutonium-processing accidents. Summaries of these two accidents follow as derived from Stratton (1967) or Paxton (1966). No criticality accidents have occurred regarding mechanical processing, storage of plutonium materials, or transportation of plutonium materials.

#### **7.3.2.1 Los Alamos Accident - December 30, 1958**

A nuclear criticality accident occurred on December 30, 1958, at the Los Alamos Scientific Laboratory, killing one worker and overexposing two other workers. The criticality occurred in a 225-gal, 38-in.-diameter stainless steel tank, with a thick organic layer containing 3.27-kg plutonium floating on a dilute aqueous solution of 60-g plutonium in 330 L. The tank was cylindrical and water-reflected. The tank contents were stirred, mixing the contents into a criticality configuration. Microbubbles, thermal expansion, and continued mixing of the tank eliminated the critical configuration. The excursion consisted of a single pulse of  $1.5 \times 10^{17}$  fissions. The operator near the tank received a lethal dose of 12,000 rem ( $\pm 50\%$ ), while two workers who assisted the operator received doses of 134 rem and 53 rem. The tank was supposed to have only 0.125 kg of plutonium; however, a gradual accumulation of solids during the 7.5-year operating history of the plant resulted in 3.27-kg plutonium in the tank.

### **7.3.2.2 Hanford-Recuplex Plant Accident - April 7, 1962**

On April 7, 1962, a criticality accident occurred at a multipurpose plutonium-recovery operation at the Recuplex Plant, Hanford, Washington. During a clean-up operation, about 46 L of solution containing 1400- to 1500-g plutonium was directed into a 69-L glass transfer tank that led to the criticality accident. The tank was spherical, 93% full, and unreflected. Solutions in the tank generally contain only a fraction of a gram per liter; however, in this situation apparently the solution was drawn from a sump through a temporary line that was being used for cleanup. The excursion had an initial pulse of about  $10^{16}$  fissions. Following this spike, the tank was supercritical for 37.5 hours with the power level steadily decreasing (Stratton, 1967). The total yield of the accident was about  $8.2 \times 10^{17}$  fissions distributed over a 37-hour time period with about 20% in the first half-hour. The excursion concluded after the boiling off of about 6 L of water and the settling of some organic matter after it had extracted plutonium from the aqueous phase. Three workers in the vicinity of the tank during the initial spike received doses greater than regulatory limits. One worker about 5 to 6 ft from the tank received 110 rem, another approximately 9 ft away received about 43 rem, and the final worker about 26 ft away received about 19 rem.

## **7.4 CRITICALITY ALARMS AND NUCLEAR ACCIDENT DOSIMETRY**

Requirements for criticality alarm systems and nuclear accident dosimetry are presented in this section. Criticality alarm systems provide rapid warning to individuals in the immediate accident location and nearby locations to evacuate to a predesignated assembly location. Specific requirements for the criticality alarm system are found in DOE Order 420.1 (DOE, 1995c) and ANSI/ANS-8.3 (ANSI, 1986a). Key requirements that may be of interest for the health physics staff are summarized in Section 7.4.1. Paxton (1966) noted that lives have been saved in past criticality accidents by radiation alarms coupled with effective evacuation procedures. Nuclear accident dosimetry, discussed in Section 7.4.2, provides the means of determining the dose to workers in the vicinity of the excursion.

### **7.4.1 Criticality Alarm System**

In accordance with DOE Order 420.1, the nuclear criticality safety program shall be evaluated and documented and shall include:

Assessment of the need for criticality accident detection devices and alarm systems, and installation of such equipment where total risk to personnel will be reduced.

The basic elements and control parameters of programs for nuclear criticality safety shall satisfy the requirements of the following American Nuclear Society's ANSI/ANS nuclear criticality safety standards:

ANSI/ANS-8.3-1986, "Criticality Accident Alarm System," however paragraphs 4.1.2, 4.2.1 and 4.2.2 shall be followed as modified in section 4.3.3.c and e of DOE Order 420.1.

Revision to the ANSI/ANS standard listed above will place the appropriate Section of DOE Order 420.1 under immediate review by DOE. Revised ANSI standards shall not be used unless an exemption is granted or it is incorporated into a DOE Order.

As specified in ANSI/ANS-8.3, the need for criticality alarm systems shall be evaluated for all activities in which the inventory of fissionable material in individual unrelated work areas exceeds 700 g of <sup>235</sup>U, 520 g of <sup>233</sup>U, 450 g of <sup>239</sup>Pu, or 450 g of any combination of these three isotopes.

-- If the fissionable material mass exceeds the ANSI/ANS-8.3 limits and the probability of criticality is greater than  $10^{-6}$  per year, a criticality alarm system shall (DOE Order 420.1, Section 4.3.3) be provided to cover occupied areas in which the expected dose exceeds 12 rad in free air. Nuclear accident dosimetry shall also be provided, as required by 10 CFR 835.1304. The criticality alarm system should include a criticality detection device and a personnel evacuation alarm.

Note: In what follows,  $10^{-6}$  per year is used as a measure of credibility, and does not mean that a probabilistic risk assessment (PRA) has to be performed.



Reasonable grounds for incredibility may be presented on the basis of commonly accepted engineering judgment.

- If the fissionable material mass exceeds the ANSI/ANS-8.3 limits and the probability of criticality is greater than  $10^{-6}$  per year, but there are no occupied areas in which the expected dose exceeds 12 rad in free air, then only a criticality detector system (i.e., nuclear accident dosimetry) is needed (DOE Order 420.1, Section 4.3.3).
- If the fissionable material mass exceeds the ANSI/ANS-8.3 limits, but a criticality accident is determined to be impossible or less than  $10^{-6}$  per year (per a safety analysis report documentation), then neither a criticality alarm system nor nuclear accident dosimetry is needed (DOE Order 420.1, Section 4.3.3).

ANSI/ANS-8.3 provides several additional requirements regarding criticality alarm systems. The alarm signal shall be for immediate evacuation purposes only and of sufficient volume and coverage to be heard in all areas that are to be evacuated. Information on sound levels of the alarm can be found in ANSI/ANS-8.3. The alarm trip point shall be set low enough to detect the minimum accident of concern. The minimum accident of concern may be assumed to deliver the equivalent of an absorbed dose in free air of 20 rad at a distance of 2 m from the reacting material within 60 sec. The alarm signal shall activate promptly (i.e., within 0.5 sec) when the dose rate at the detectors equals or exceeds a value equivalent to 20 rad/min at 2 m from the reacting material. A visible or audible warning signal shall be provided at a normally occupied location to indicate system malfunction or loss of primary power. Each alarm system should be tested at least once every three months. An evacuation drill shall be conducted at least annually.

Criticality alarm systems may consist of one to several detectors per unit. In multi-detector units (e.g., three detectors), at least two detectors shall be at the alarm level before initiating the alarm; in redundant systems, failure of any single channel shall be into the trip state (ANSI, 1986a).

#### **7.4.2 Nuclear Accident Dosimetry**

In accordance with DOE Order 420.1, the nuclear criticality safety program shall be evaluated and documented and shall include:

Assessment of the need for criticality accident detection devices and installation of such equipment where total risk to personnel will be reduced.

Nuclear accident dosimetry is required when the fissionable material mass exceeds the ANSI/ANS-8.3 limits discussed in Section 7.4.1 and the probability of criticality is greater than  $10^{-6}$  per year.

Requirements for nuclear accident dosimetry programs at DOE facilities are found in 10 CFR 835.1304 (DOE, 1993c). A nuclear accident dosimetry program shall include the following:

- A method to conduct initial screening of personnel involved in a nuclear accident to determine whether significant exposures to radiation occurred;
- methods and equipment for analysis of biological materials;
- a system of fixed nuclear accident dosimeter units (sometimes referred to as area dosimeters); and
- personnel nuclear accident dosimeters (PNADs) worn by all personnel who enter locations in which installed criticality alarm systems are required.

Additional desirable features of a nuclear accident dosimetry program include:

- Facilities to evaluate fixed dosimeters and/or PNADs;
- a method to determine the approximate neutron spectrum;
- a method to determine the activity of  $^{24}\text{Na}$  in blood and  $^{32}\text{P}$  in hair; and
- a method to correct dosimeter results for actual spectrum (if known).

#### **7.4.2.1 Initial Screening Evaluation**

A nuclear accident dosimetry program should provide absorbed dose information within 24 hours after the incident. A nuclear accident dosimetry program shall include a method to conduct initial screening of personnel involved in a nuclear accident to determine whether significant exposures to radiation have occurred (10 CFR 835.1304)[also see ANSI N13.3 (ANSI, 1969b)]. Discussions on initial screening evaluations to segregate exposed from unexposed individuals (sometimes referred to as "quick sort techniques") are found in several references (Moe, 1988; Delafield, 1988; Petersen and Langham, 1966; Hankins, 1979; Swaja and Oyan, 1987).

A common initial screening method is to provide all workers in areas requiring nuclear accident dosimetry with an indium foil in their personnel dosimeter or security badge. During a criticality excursion the foil will become activated by neutrons per the  $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$  reaction and can be measured with a portable beta-gamma survey instrument or ion chamber. The  $^{116\text{m}}\text{In}$  has a 54-min half-life and releases a 1-MeV beta (maximum energy) and a 1.3-MeV gamma (80% of the time).

An alternate screening is to measure body activity due to neutron activation of the sodium in the blood via the  $^{23}\text{Na}(n, \gamma)^{24}\text{Na}$  reaction. Sodium-24 has 15-hour half-life and releases a 1.4-MeV beta (maximum energy) and two gammas (1.37 MeV and 2.75 MeV). A beta-gamma survey meter is used to

measure the  $^{24}\text{Na}$  activity in the blood by placing the detector probe against the individual's abdomen and having the individual bend forward to enclose the detector (Moe, 1988). Alternatively, the probe can be positioned under the armpit with the open window facing the chest area. Moe (1988) noted that this method is less sensitive than the use of indium foils and even a small reading can indicate a significant exposure. An approximate equation to calculate worker dose (D) based on body weight and instrument reading is shown in Equation 7.1:

$$D \text{ (Gy)} = \frac{80 \text{ (instrument reading in mR / h)}}{\text{Body weight (lb)}} \quad (7.1)$$

Differences in incident neutron energy spectrum, orientation, and measurement techniques relative to conditions used to develop activity-dose correlations can cause significant errors in estimated radiation dose based on quick-sort surveys. Swaja and Oyan (1987) showed that radiation doses estimated from induced body activity can vary by a factor of about 2 due to neutron energy spectrum or orientation effects and by as much as 30% due to probe position. Doses based on indium foil activity can vary by a factor of about 9 due to neutron energy spectrum effects, a factor of 3 depending on foil orientation relative to the incident field, and a factor of about 2 due to probe window setting. Swaja and Oyan (1987) recommended that those count rates above background during quick-sort techniques should be initially interpreted only as an indication that the person has been exposed.

#### **7.4.2.2 Fixed and Personnel Nuclear Accident Dosimeters**

A comprehensive nuclear criticality dosimetry system should consist of stationary (fixed-location, area) dosimeters, neutron and gamma dosimeters worn by personnel (i.e., PNADs), and specialized laboratory equipment to evaluate the dosimeters.

Fixed nuclear accident dosimeter units should be capable of determining neutron doses in the range of 10 rad to 10,000 rad with an accuracy of  $\pm 25\%$ . They

should also be capable of providing the approximate neutron spectrum to permit the conversion of rad to rem. The gamma-measuring component of the dosimeter should be capable of measuring doses in the range of 10 rem to 10,000 rem in the presence of neutrons with an accuracy of about  $\pm 20\%$ . The number of fixed dosimeter units needed and their placement will depend on the nature of the operation, structural design of the facility, and accessibility of areas to personnel. Generally, dosimeters should be placed such that there is as little intervening shielding and as few obstructions as possible (ANSI, 1969b). The number and placement of dosimeters should be periodically reverified to reflect changes in building design and operations. Ease of dosimeter recovery after a criticality event should be considered in their placement, including the possible need for remote retrieval.

10 CFR 835.1304 requires that PNADs be worn by all personnel who enter a controlled area with locations requiring an installed criticality alarm system. The PNADs should be capable of determining gamma dose from 10 rad to 1000 rad with an accuracy of  $\pm 20\%$  and neutron dose from 1 rad to 1000 rad with an accuracy of  $\pm 30\%$  without dependence upon fixed-unit data.

ANSI N13.3 (ANSI, 1969b) provides general criteria for nuclear accident dosimeters that are reviewed below. Dosimeters, both fixed and personnel, should be protected against radioactive contamination to avoid false measurements. Periodic inventory methods should be established and audits made to ensure that the dosimeters are not removed or relocated without appropriate approvals. Techniques for estimating the effect of body orientation at the time of the exposure should also be developed.

**Neutron-Measuring Component of Dosimeter.** Criticality accidents create a wide range of neutron energies. Since the neutron dose per unit fluence is strongly dependent on neutron energy, knowledge of the neutron energy spectrum is important in accident dosimetry. In criticality accidents, neutrons with energies greater than about 100 keV contribute most of the dose; therefore, measurement of the fast neutron dose is of most importance. See Delafield

(1988) for a review of the different types of neutron dosimeters available for accidents.

**Gamma-Measuring Component of Dosimeter.** Delafield (1988) noted that the ratio of the gamma rays to neutron dose will vary according to the type of critical assembly and whether or not additional shielding is present. For unshielded assemblies, the gamma-to-neutron ratio can range from 0.1 for a small heavy metal system up to about 3 for a small hydrogen-moderated solution system. A concrete or hydrogenous shielding material will increase the gamma-to-neutron ratio. Gamma dose can be determined by TLD, film, or radiophotoluminescent glass.

**Dosimeter Comparison Studies.** Sims and Dickson (1979) and Sims (1989) present a summary of nuclear accident dosimetry intercomparison studies performed at the Oak Ridge National Laboratory Health Physics Research Reactor. The most recent summary (Sims, 1989) showed that of the 22 studies conducted over 21 years, 68% of the neutron dosimeter results were within the  $\pm 25\%$  accuracy standard and 52% of the gamma dosimeter results were within the  $\pm 20\%$  accuracy standard. Most measurements that failed to meet the accuracy standards overestimated the actual dose. Some of their other findings include the following:

- Doses from hard neutron energy spectra are more accurately measured than those from soft energy spectra
- The threshold detector unit (TDU) is the most accurate type of nuclear Accident neutron dosimeter; however, its use is declining due to increasingly strict control of small quantities of fissionable materials
- Activation foils (ACT) are the most popular nuclear accident neutron dosimeter
- For gamma dosimeters, TLDs are the most popular and the least accurate, and film is the least popular and the most accurate.

#### **7.4.2.3 Biological Indicators**

Earlier in this section, a quick-sort method was described using neutron activation of sodium in the blood as an indicator of worker exposure. More sophisticated laboratory analysis of blood samples can be performed to obtain a more accurate estimate of worker dose, as discussed in Delafield (1988) and Hankins (1979). The use of neutron activation of sulfur in hair ( $^{32}\text{S}(\text{n}, \text{p})^{32}\text{P}$ ) is another method to estimate absorbed dose for workers involved in a criticality accident (Petersen and Langham, 1966). The orientation of the subject can also be determined by taking samples of hair from the front and back of the person. Hankins (1979) described a technique for determining neutron dose to within  $\pm 20\text{-}30\%$  using a combination of blood and hair activations. Their evaluation was independent of the worker's orientation, of shielding provided by wall and equipment, and of neutron leakage spectra.

### **7.5 RESPONSIBILITIES OF HEALTH PHYSICS STAFF**

The health physics staff should have a basic understanding of program structure, engineering criteria, and administrative controls as related to nuclear criticality safety as reviewed in earlier sections of this chapter. Additionally, the health physicist's responsibilities include emergency instrumentation and emergency response actions.

#### **7.5.1 Routine Operations**

During routine operations the health physics staff's responsibilities related to nuclear criticality safety include calibrating, repairing, and maintaining the neutron criticality alarm detectors and nuclear accident dosimeters, and maintaining appropriate records. The health physics staff should be knowledgeable of criticality alarm systems, including alarm design parameters, types of detectors, detector area coverage, alarm set-points, and basic control design. The staff should also be familiar with plans for emergency response.

The health physics staff should maintain an adequate monitoring capability for a nuclear criticality accident. In addition to the criticality alarm systems and the fixed nuclear accident dosimeters discussed above, remotely operated high-range gamma instruments, personal alarming dosimeters for engineering response/rescue teams, neutron-monitoring instrumentation (in case of a sustained low-power critical reaction), and an air-sampling capability for fission gases should be maintained.

Other support activities may include assisting the nuclear criticality safety engineer or operations staff in performing radiation surveys to identify residual fissionable materials remaining in process system or ventilation ducts.

### **7.5.2 Emergency Response Actions**

The priorities of line management (which could include involving the health physics staff) during a criticality event should be to rescue personnel, prevent further incidents or exposures, and quickly determine those who have been seriously exposed (Moe, 1988). To support these emergency response actions, the health physics staff should be trained in facility emergency procedures. These emergency procedures include evacuation routes, personnel assembly areas, personnel accountability, care and treatment of injured and exposed persons, a means for immediate identification of exposed individuals, instrumentation for determining the radiation levels at the assembly area, and the re-entry and formation of response teams.

Emergency response procedures for conducting the initial quick sort of workers should specify measurement techniques and require that surveyors record methods and instrument settings used for quick-sort operations to ensure proper interpretation of the results. Field results should be compared to pre-established activity-dose relationships developed as part of emergency response procedures to determine if a worker was exposed. Other indicators such as a discharged self-reading dosimeter could also be an indication of a possible exposure.

As an immediate follow-up action for workers identified as being exposed during a quick-sort procedure, a more accurate dose estimate should be made using PNADs, fixed-location accident dosimeters, or biological activity analyses ( $^{24}\text{Na}$  in the blood or  $^{32}\text{P}$  in



the hair). Part of these more accurate analyses should include: 1) better definition of source characteristics, 2) location of moderating materials, and 3) location and orientation of the person(s) at the time of exposure and action of the person following the irradiation. The health physics staff can provide valuable information to support this analysis, particularly regarding the location and orientation of workers to the excursion if they are involved in the rescue and initial monitoring procedures.

Health physics staff will be responsible for retrieving fixed nuclear accident dosimeters and ensuring that PNADs from any exposed workers are submitted for analysis.

### **7.5.3 Special Considerations During Decommissioning Activities**

Before decommissioning or disposal of any facilities or equipment, an evaluation should be performed to assess the potential holdup of fissionable material in any equipment. These types of measurements may require the assistance of health physics staff.

Some strippable coatings and surface fixing films are good neutron moderators. Nuclear criticality safety specialists should be consulted when using these coatings to decontaminate surfaces because criticality could be a concern, depending on the geometry of the removed coating when in the disposal unit.

## **7.6 DEPARTMENT OF ENERGY PLUTONIUM VULNERABILITY ANALYSIS STUDY**

In March 1994, Department of Energy Secretary Hazel R. O'Leary commissioned a comprehensive assessment to identify and prioritize the environment, safety and health vulnerabilities that arise from the storage of plutonium in the DOE facilities and determine which are the most dangerous and urgent. These vulnerabilities are degradation in plutonium materials and packaging, and weakness in facilities and administrative controls that can expose workers and public, or contaminate the environment. The summary of the results presented in this section is taken from DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

The assessment was commissioned because of recent ruptures of stored plutonium packages and the need to store safely the large amount of plutonium-bearing materials held by the DOE in its

aging facilities. The ultimate goal of the assessment was to facilitate safe and stable interim storage until its final disposition, which is not expected to take place for at least 10 to 20 years. The assessment covered 166 facilities at 35 site and employed a Working Group process. The Plutonium Working Group combined the talent of DOE federal staff, site management and operations contractors, consultants and stakeholders. The Working Group developed plans and technical approaches for the assessment and evaluated the assessment results. Overall, this assessment took more than six months and 80,000 person-hours.

During the assessment, the DOE discussed information about vulnerabilities with stakeholders. About 45 stakeholder groups were involved in either the Working Group meetings or local activities associated with site assessments.

Excluding the classified mass of plutonium contained in nuclear weapon pits at the Pantex Plant in Texas, these sites hold 26 metric tons of plutonium. Most of this is located in Rocky Flats, Colorado; Hanford, Washington; Argonne-West, Idaho; Los Alamos, New Mexico; and Savannah River Site, South Carolina. The report details the most significant vulnerabilities within each site and across all sites. The Working Group categorized and classified vulnerabilities based on possible effects on workers, the public or environment.

The DOE-wide assessment identified 299 environment, safety and health vulnerabilities at 13 sites, consisting of 91 material/packaging vulnerabilities, 140 facility condition vulnerabilities and 68 institutional vulnerabilities.

In general, the vulnerabilities identified in this assessment pose the greatest hazards to workers. Packaging, which the Working Group found to be widely deficient for long term storage, is often the only barrier that separates the workers from the plutonium. Plutonium solutions are the form most difficult to store and present unique hazards. Plutonium scrap and residue forms are reactive, and some are corrosive enough to degrade containers. Plutonium metals and oxides generally present fewer problems, but much of this material is stored in plastic, which can react with plutonium and cause container failure.

Facility conditions that cause vulnerabilities include aging safety systems, holdup of plutonium in process systems, and design problems that weaken the ability to mitigate accidents like fires or earthquakes. In addition to their impact on workers, such large-scale events have the potential to

release plutonium that could effect the public and environment. Institutional vulnerabilities involve incomplete safety analyses, loss of experienced staff, and operational problems such as a backlog of maintenance items on systems that are important to safety.

The assessment found Rocky Flats Buildings 771 and 776 to be the most vulnerable facilities, based on combinations of their vulnerabilities and amount of plutonium they hold. These buildings are more than 35 years old and have design deficiencies. The next group of most vulnerable facilities are the Savannah River Site's Building 235-F, FB-Line and Old HB-Line; Hanford's Plutonium Finishing Plant; and Rocky Flats Building 779, 707, and 371. The material in these facilities includes plutonium solutions and reactive materials.

This assessment provided the information base that will improve the Department's plan for safely managing the future disposition of its plutonium. While most vulnerabilities were already known, this assessment improved DOE's understanding of the issues. It has also enabled the Department to document vulnerabilities, identify new ones and set priorities which will establish a systematic approach to corrective action. DOE began formulating corrective action plans to achieve safe and stable interim storage in September 1994.

The assessment reached several conclusions. Plutonium package failures and facility degradation will increase in the future unless problems are addressed in an aggressive manner. The Department needs a strong, centrally coordinated program to achieve safe interim storage of plutonium. Priority must be given to plutonium solutions, chemically reactive scrap/residues and packaging with plastics or other organic compounds. Much of the Department's plutonium inventory, including plutonium in holdup, must be better characterized and site-specific programs must be implemented to establish package design lives. Management priorities at some site should be reassessed to provide proper attention to those facilities identified as most vulnerable by this assessment. Sites must evaluate institutional vulnerabilities such as the loss qualified staff, and compensate for them. Standards or guidelines for packaging, storage and surveillance of plutonium scrap/residues and solutions must be developed and implemented. Finally, the Department and its stakeholders have just begun to work together to clean up the remnants of weapons production processes.

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## **8.0 WASTE MANAGEMENT**

A material is a waste once there is no identified use or recycle value for it. Normally, wastes are considered by their physical form as either solids, liquids, or gasses, except that containerized liquids are considered solid waste under some of the current regulations. Although these forms are each processed differently, there are interrelationships. For example, it may be possible to reduce solid waste by replacing disposable protective clothing with reusable clothing that must be laundered. The laundry will produce liquid waste. In treating liquid waste, solids may be generated, for example, filters or ion exchange resins. By careful engineering, waste generation, and treatment alternatives, a site can minimize the total waste volume and elect to generate types of waste that can be disposed of. The following sections address potentially contaminated waste and waste terminology and handling of airborne waste, solid waste, and liquid waste. The treatment of excess materials to reclaim plutonium is not a waste treatment process and is not discussed here.

### **8.1 POTENTIALLY CONTAMINATED WASTES**

This section discusses the generation, processing, storage, and disposal of wastes in plutonium facilities. It is divided by waste types, treatability groups, and waste disposal.

#### **8.1.1 Waste Types**

In addition to the classification of waste by physical form, regulatory definitions determine how waste can be disposed. The Secretary of Energy Notice 37-92, "Waste Minimization Crosscut Plan Implementation" (SEN, 1992), requires annual reports of waste generation by type, waste stream, site, and program. The waste classifications used in the DOE Annual Reports are defined in Table 8.1.

A plutonium facility may generate any of these types of waste, except that high-level waste (HLW) will be generated only from irradiated reactor fuel. Any waste containing at least 100 nCi/g of transuranics (TRU), including plutonium, will be classified as TRU or TRU mixed waste. Waste containing detectable quantities of radioactive materials but less than 100 nCi/g of transuranics will be low-level waste (LLW).

The distinction between sanitary waste and very low-level radioactive waste can be technically a difficult one. Sometimes, material is designated LLW waste because the conditions of use could have resulted in contamination that would be difficult to detect. Techniques and limitations for doing this are discussed below with reference to solid waste.

### **8.1.2 Treatability Groups**

In addition to being classified by type, as discussed above, wastes are classified by treatability group, depending on the treatment the waste receives. The common treatability groups are defined in Table 8.2. These are reported in each site's annual waste management report.

<b>Table 8.1. Waste Types<sup>(a)</sup></b>	
<b>HLW</b>	High-level waste (HLW) is the material that remains following the reprocessing of spent nuclear fuel and irradiated targets from reactors. The HLW is highly radioactive and generates heat on its own. Some of its elements will remain radioactive for thousands of years. Because of this, HLW must be managed very carefully and all handling must be performed from behind heavy protective shielding.
<b>LLW</b>	Low-level waste (LLW) is any radioactive waste that is not HLW, spent nuclear fuel, TRU waste, or uranium mill tailings. The LLW is typically contaminated with small amounts of radioactivity dispensed in large amounts of material. The LLW is generated in every process involving radioactive materials in the DOE including decontamination and decommissioning projects.
<b>MW</b>	Mixed waste (MW) is waste that contains both radioactive and hazardous wastes. Any of the types of radioactive waste described can be a mixed waste if it contains any hazardous wastes. In fact, all of DOE's HLW is mixed waste because of the chemicals used to reprocess the fuel that resulted in the generation of the material or because it is suspected to contain hazardous materials.
<b>TRU</b>	Transuranic (TRU) waste refers to waste materials containing elements with atomic numbers greater than 92. These elements are generally alpha-emitting radionuclides that decay slowly. The TRU waste contains a concentration of these elements greater than 100 nCi/g. The TRU waste is not as intensely radioactive as HLW. The TRU waste also decays slowly, requiring long-term isolation.
<b>Sanitary Waste</b>	Sanitary waste is waste that is neither hazardous nor radioactive.
<b>Hazardous Waste</b>	Because of its quantity, concentration, and physical, chemical, or infectious characteristics, hazardous waste may cause or significantly contribute to an increase in mortality, or an increase in serious irreversible or incapacitating reversible illness; it may pose a potential hazard to human health or the environment when improperly treated, stored, transported, disposed of, or otherwise managed.

<b>Table 8.1. Waste Types<sup>(a)</sup> (continued)</b>	
<b>RCRA (USC, 1976a) Regulated Waste</b>	Solid waste, not specifically excluded from regulation under 40 CFR 261.4 (EPA, 1994a), or delisted by petition, that is, either a listed hazardous waste (see 40 CFR 261.30 - 261.33) or waste exhibiting hazardous characteristics.
<b>State Regulated Waste</b>	Any other hazardous waste not specifically regulated under TSCA or RCRA, which may be regulated by a State or local authority. An example of such waste is used oil.
<b>TSCA (USC, 1976b) Regulated Waste</b>	Hazardous chemical wastes, both liquid and solid, containing more than 50 parts per million of polychlorinated byphenyls.
(a) Definitions from DOE/S-0101, <i>U.S. DOE Annual Report on Waste Generation and Waste Minimization Progress, 1991-1992</i> , February 1994 (DOE, 1994o).	

<b>Table 8.2. Treatability Groups</b>
<b>LLW</b>
Contact-handled LLW: exposure rate of 200 mR/h or less on contact Remote-handled LLW: exposure rate greater than 200 mR/h on contact
<b>TRU</b>
Contact-handled TRU: exposure rate of 200 mR/h or less on contact Remote-handled TRU: exposure rate greater than 200 mR/h on contact
<b>Mixed Waste (MLLW and MTRU)</b>
Treatable mixed waste has an existing treatment that will eliminate or encapsulate (TCLP) the hazardous constituents of the mixed waste, rendering it LLW or TRU. Treatable includes treatment of mixed waste that results in volume reduction.  Non-treatable: no treatment exists

Plutonium facilities generate mostly contact-handled TRU waste (even though they may do much of the handling and processing within glove boxes for contamination control). The most common treatment will be compaction although some facilities have incinerators available.

Depending on the treatment methods available, waste streams may be tailored to be amenable to treatment. Some facilities are able to incinerate TRU waste. Facilities with this capability may need to eliminate halogenated, nitrogenated, or sulfur-containing



materials to maximize incinerator acceptance and minimize hazardous effluents from the incinerator.

### **8.1.3 Waste Disposal**

Waste classifications and treatability groups are important because they determine waste disposal options.

**Sanitary waste** is by far the least costly and easiest to dispose of. Liquid sanitary waste is disposed of in sanitary sewerage systems or septic systems. Sanitary solid waste is nearly always disposed of by landfill disposal or by incineration with landfill disposal of ash. Because sanitary waste disposal facilities still face various siting and permitting requirements, it is desirable to minimize waste volumes.

**Hazardous waste** is second in ease of disposal for most DOE facilities. Hazardous waste can be treated to eliminate the hazard only if a permit for the particular waste stream has been granted by the Environmental Protection Agency (EPA). Hazardous waste treatments permitted in DOE facilities are usually limited to pH adjustment, precipitation, and ion exchange for liquid waste and compaction or incineration for solid waste. Combustible liquids may be incinerated either onsite or offsite, as conditions permit.

**Low-level waste** is still disposable at most sites. For NRC and state-licensed facilities, commercial disposal is an option, but subject to the requirements of the Low-Level Waste Policy Amendments Act (USC, 1985), which requires individual states or groups of states, called compacts, to develop local disposal facilities. In general, local facilities have not been developed, so disposal volumes are severely limited and/or significant surcharges are imposed in addition to the already high disposal cost.

Several DOE sites are currently permitted to dispose of their own low-level waste by burial. Other DOE sites have long-term storage facilities. In some cases, DOE waste is being placed in retrievable storage in the hopes that the classification of the facility can be changed and the waste allowed to remain permanently.

**Mixed waste** disposal facilities require all of the permitting for radioactive waste disposal facilities plus all of the permitting for hazardous waste disposal facilities. For this reason, there are very few such facilities in operation, and in general they are rather restricted in the type of waste they can accept. If possible, it is generally better to treat the waste than to destroy or chemically alter the hazardous component. In some cases, mixed waste may be treated to encapsulate the hazardous component so that it no longer has the leachability or other characteristics that cause it to exhibit hazardous properties. Mixed waste requires special permits for treatment, so it is generally preferable to avoid generating it or to treat it in connection with some other process while it is a useful material (before it becomes a waste). For example, if the hazardous component is a metal with some recycle value, or if there are recycle metals in the material, it may be best to alter the process to plate or precipitate the material as a final step in the process line, before it is declared a waste.

Most plutonium facilities will produce **TRU waste** or **TRU mixed waste**. According to national policy, DOE TRU waste is supposed to be permanently disposed of at the Waste Isolation Pilot Plant (WIPP) under construction and testing at Carlsbad, New Mexico. Volume allocations have been given to each DOE site for the waste to be placed there, so limiting the quantity of high-level TRU waste is extremely important. The current date at which WIPP will be permitted to accept waste and the cost of waste disposal there have not yet been determined. In the interim, virtually all DOE facilities are required to store TRU on the site where it is generated.

Therefore, volume reduction of TRU waste is highly desirable. Incineration offers the greatest volume reduction and has the added advantage of destroying some types of hazardous constituents (flammable and other organic compounds).

**High-level wastes** are slated to be disposed of at a high-level waste repository. A site at Yucca Mountain, Nevada, is being evaluated for this purpose. In the interim, TRU waste is being stored either at the sites that are generating it or, for some DOE facilities, at the Nevada Test Site, until a final repository is available. Long-term maintenance of interim storage facilities, plus the prospect of later moves to the final disposal site and burial at that site, make high-level waste very costly.

## **8.2 AIRBORNE WASTE**

The only airborne plutonium likely to arise from either normal operations or decommissioning of DOE facilities will be in a particulate form. Although plutonium vapors are possible during cutting and perhaps some grinding operations, they will soon condense to particulate material.

### **8.2.1 Design Objectives**

Plutonium particulates are notoriously difficult to confine and extensive use is made of glove boxes, local ventilation systems, fixatives, and other means to minimize generation of particulates and to confine them. The high-efficiency particulate air (HEPA) filter is the backbone of plutonium air-cleaning systems. Such filters are certified to have a 99.93% removal efficiency for particulates of 0.3  $\mu\text{m}$  and larger and are normally used with at least two in series.

Because confinement systems are subject to component failures and other accidents, differential air pressures are normally maintained so that a breach of containment will not affect occupied areas or the environment. Glove-box lines are at the lowest pressure, plutonium laboratories at a higher pressure, and other occupied areas at the highest pressure but still negative with respect to the outside.

Because plutonium air-cleaning systems are usually expensive to service (requiring workers to be dressed in multiple layers of protective clothing and respiratory protection), and plutonium waste is expensive to dispose of, measures are taken to protect the life of plutonium air-cleaning systems. Extraneous particulates are eliminated by HEPA filtration of incoming air. (These HEPA filters may be disposed of as sanitary waste.) Roughing pre-filters are used to capture the bulk of particulates and prolong the life of HEPA filters.

Care must be taken in designing HEPA filter installations for plutonium facilities so that provisions are made to safely change the filters while maintaining contamination control. Such measures normally include redundant banks of filters (in parallel) that can be valved out for filter change, location of HEPA filter banks in enclosed rooms that are themselves HEPA-filtered, and appropriate provisions for filter bag-out.

New filters must be tested after they are installed to ensure proper gasketing, etc. Once in place, they must be periodically retested to ensure that HEPA efficiency is maintained. For this reason, HEPA filter installations must have ports for the introduction of a challenge aerosol upstream of the filter and collection of a representative sample in a region of laminar flow downstream of the filter. The HEPA filters in plutonium use sometimes fail from mechanical fatigue and vibration rather than plugging or being subject to some other mechanical failure. The proper design of HEPA filtration systems and proper sampling provisions are discussed in DOE Order 6430.1A, General Design Criteria (DOE, 1989b); ANSI N510-1989, Testing of Nuclear Air-Cleaning Systems (ANSI, 1989d); ANSI/UL 586-1990, High-Efficiency Particulate Air Filter Units (ANSI, 1990); ANSI/ASME N509-1989, Nuclear Power Plant Air Cleaning Units and Components (ANSI, 1989b); DOE Implementation Guide. Workplace Air Monitoring (DOE, 1994g); and ANSI N13.1-1969, Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities (ANSI, 1969b).

In addition to the above features of the air-handling system, there may be process-selection features that will minimize the generation of airborne plutonium. If at all possible, plutonium compounds should be handled in sealed containers or, in the case of a metallic solid, the material encapsulated. Wet mechanical processes, such as cutting and grinding, usually generate fewer particulates than dry ones, so they are often preferred. However, it is also important to minimize the use of chemicals that will attack the air-cleaning system or contaminate the filters with hazardous chemicals, making them mixed waste. Even moisture will shorten the life of HEPA filters, so wet processes should be enclosed to the extent practicable and demisters and/or heaters used to pretreat the air from wet processes prior to HEPA filtration.

The final consideration in the design of air cleaning systems for plutonium operations is the probability and consequences of accidents. In general, plutonium air-handling systems are designed so that all probable accidents, including the failure of a single HEPA filter, do not have measurable consequences offsite. It will be necessary to design the system for all probable meteorological conditions, including (for some regions of the country) tornados. The system must also be designed so that some improbable (but not impossible) events (accidents) have consequences that are less than catastrophic. For example, the simultaneous failure of two HEPA filters in series is highly unlikely

(without a common cause such as high differential pressure from an explosion or meteorological event) but facilities must be designed so that these events are not likely to cause fatalities offsite. The minimum performance criteria for the air-cleaning systems are dictated by DOE design criteria. Other design parameters are finalized during the Environmental Impact and Safety Analysis processes. They will differ from facility to facility.

### **8.2.2 Operational Controls**

Plutonium air-handling systems must be operated within the design safety envelope of the system, designated by Operational Safety Requirements (OSRs). Beyond that, there are measures that can further reduce the potential for airborne plutonium, even in glovebox operations. Even within glove boxes, plutonium should be containerized, preferably doubly encapsulated whenever possible. Spills should be cleaned up promptly. If rags or tissues are contaminated, they should be bagged as soon as possible.

### **8.2.3 Waste Treatments**

The principal treatment for cleaning plutonium from air is HEPA filtration. There are other technologies that can be used for pretreatment, but the most common is filtration. Electrostatic precipitation, wet scrubbing, demisters to remove moisture, and other technologies may have specific applications. (Treatment of the HEPA filters, a solid waste, and the wet scrubber effluent, a liquid waste, are discussed in Sections 8.3 and 8.4, respectively.)

### **8.2.4 Sampling and Monitoring**

Sampling is the primary method used to achieve a complete and accurate legal record of releases after they have occurred. The primary requirement for a particulate air sample of any type is that it be representative of the stream being measured. This translates into isokinetic sampling in a laminar flow section of the exhaust duct. The parameters needed to achieve such a sample are given in numerous references such as the DOE Implementation Guide. Workplace Air Monitoring (DOE, 1994g), and ANSI N13.1-1969, Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities (ANSI,

1969b). For sampling, the analytical methods are the same as those discussed in Chapter 3 of this document for workplace sampling.

Monitoring is used to determine if current conditions are within expected parameters and to initiate corrective action if they are not. For monitoring, the system design should conform to ANSI N42.18, Specification and Performance of On-site Instrumentation for Continuously Monitoring of Radionuclides in Effluents (ANSI, 1974b). The choice of the filter medium will depend on the analysis that will be done on the sample. For samples containing only plutonium particulate, a non-absorbing filter such as a membrane filter will have the highest efficiency for alpha counting. In all cases, the final count must be done after any residual radon has decayed because it will often result in a large amount of alpha on the filter that is not plutonium. If there are other radionuclides in the waste stream that cannot be decayed in a reasonable time, either alpha spectroscopy or chemical separation must be done. Chemical analysis must also be done if there are stable contaminants of interest such as beryllium or heavy metals. The nature of these procedures is beyond the scope of this document.

#### **8.2.5 Disposal**

Airborne effluents are not stored. Disposal of the airborne effluent, possibly containing traces of plutonium, is generally arranged by the design of the facility and the existing air-quality permits. Normally, the design of the facility is such that the method of disposal of the cleaned effluent should be unimportant during normal operation. However, the facilities are designed to minimize the impact of a filter failure or operational difficulty that results in a release. Disposal of airborne effluents is handled at the design, environmental impact assessment, and safety analysis stages of facility construction. Disposal of secondary waste from air cleaning is covered in the sections that follow.

### **8.3 SOLID WASTE**

Solid waste will come from all phases of operation and from decommissioning of plutonium facilities. Because most plutonium solid waste will be TRU (containing more than 100 nCi/g), disposal in the near future is uncertain. Thus, it is highly desirable to minimize the generation of solid waste in the design, operation, maintenance, and decommissioning of plutonium facilities.

#### **8.3.1 Design Objectives**

One of the principal means of minimizing solid waste is to minimize the area that becomes contaminated by plutonium and to ensure that all surfaces contaminated by plutonium are readily cleanable.

Glove boxes are often used to contain contamination and permit work in minimal protective clothing that can be reused to minimize waste volumes. By assuring that these are in isolated areas that are covered with easily cleanable materials and maintained at negative pressure with respect to the rest of the facility, waste is minimized even during minor accidents.

The choice of surface materials is extremely critical. For example, concrete floors will become impregnated by plutonium particulates or solutions and will require fixatives or scabbing to control contamination. Relatively large quantities of solid waste will be generated when facilities are decommissioned or major modifications are done.

Conversely, electropolished stainless steel is easily cleaned, even to releasable levels generating only small quantities of TRU waste.

Choosing components that can be easily maintained rather than totally replaced may also be an effective strategy at minimizing waste. Whenever possible, choose equipment for which high-maintenance components can be located outside of contaminated areas. For example, many mixers, saws, and other such components have been adapted so that the motor is located outside the glove box where it can be maintained or replaced without concern for contamination status, while the working or tool end operates in a contaminated environment.

### **8.3.2 Operational Controls**

Operational controls for waste-management purposes in plutonium facilities serve two distinct purposes: waste volume reduction (waste minimization) and waste classification control. Each of these is discussed briefly below. Operational controls to reduce the probability of accidents or minimize their consequences are also important but are not directly addressed as part of waste management.

#### **8.3.2.1 Waste Minimization**

Plutonium facilities should have a waste minimization program. The objective of a waste minimization program is the cost-effective reduction in the generation and disposal of hazardous, radioactive, and mixed waste. The preferred method is to reduce the total volume and/or toxicity of hazardous waste generated at the source, which minimizes the volume and complexity for waste disposal.

The waste minimization program applies to all present and future activities of the facilities that generate hazardous, radioactive, and/or mixed wastes.

Furthermore, waste minimization is to be considered for all future programs and projects in the design stages, and should be included in all maintenance and/or construction contracts.

All managers of facilities or activities that generate hazardous, radioactive, and mixed waste are responsible for:

- Minimizing the volume and toxicity of all radioactive, hazardous, and radioactive mixed waste generated, to the extent economically practicable
- preparing and updating waste minimization plans for their waste-generating facilities or activities. Small waste generators in a larger facility may be grouped with others in a facility or activity plan



*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- implementing the facility-specific or activity-specific waste minimization plan
- providing input to the organization responsible for waste characterization and minimization, to support the waste minimization program
- communicating waste minimization plans to their employees, and ensuring that employees receive appropriate training
- ensuring that existing system/equipment replacement or modification is designed and installed to minimize generation of waste
- developing new waste minimization strategies, and identifying cognizant staff for waste minimization communications between facility personnel
- identifying new waste generating facilities or activities and significant process changes to existing facilities or activities to the waste characterization and waste minimization organization.

Waste volume control, or waste minimization, involves limiting the amount of material that becomes contaminated, segregating clean and contaminated material, and prolonging the useful life of equipment and material to minimize replacement. Sometimes, materials can be completely cleaned so that disposal as sanitary waste (or refurbishment in clean areas) is an option.

Program design decisions can affect TRU waste-generation. For example, the quantity of protective clothing may be a significant factor. If an incinerator is available, combustible protective clothing may be selected to have a low ash content and generate a minimum of harmful effluents such as oxides of nitrogen or halogenated compounds. In other facilities, water-washable, reusable protective clothing may minimize waste disposal.

In many nuclear facilities, contamination of packaging materials is a problem. For example, if a tool or material (e.g., a pump or some ion exchange resin) is to

be used in a contaminated area, as much of the packaging material must be removed as possible before the material enters the radiological area.

Another opportunity for waste minimization occurs when materials are used as a contingency protection against contamination. For example, strippable coatings may be applied to an area that is not expected to become contaminated or may receive only minor contamination so that it can be easily cleaned. Another example involves the disposition of disposable surgeons' gloves, which are routinely worn inside glove-box gloves. Unless there are serious contamination control problems in the facility, these can be surveyed and disposed of as sanitary waste rather than LLW or TRU waste.

If a piece of equipment is to have more than a single use in a contaminated environment, every possible measure should be taken to ensure its continued reliability rather than relying on frequent replacements. Tools should be of the highest quality and maximum flexibility consistent with the situation. For example, if a wrench is needed to maintain a piece of equipment in a glove-box, consideration should be given to future needs and storage provisions. A socket set with interchangeable sockets may ultimately create less waste than a box-end wrench of each size that is needed.

Likewise, all tools and equipment to be placed in a contaminated environment should be tested for reliability and preferably used on a clean mock-up to ensure their serviceability before they become contaminated. There is often a temptation to put the equipment into the plutonium service when it first arrives rather than test it completely first. This can result in unnecessary waste volume.

#### **8.3.2.2 Waste Classification Control**

Many operational controls involve measures to ensure that the waste generated is TRU waste rather than mixed-TRU waste, or that if it is mixed-TRU, it is of a composition that can be treated. Tight controls in the following areas are necessary to minimize mixed waste (and hazardous waste) problems: procurement of hazardous chemicals, actions of subcontractors and vendors, and

training of workers. In some cases, decontamination processes have been used that result in mixed waste, such as Freon cleaning, electropolishing, and chemical decontamination. These should be used only after due consideration of the waste-management consequences. In some cases, these mixed wastes can be readily treated; in other cases, their use needs to be avoided. Some new techniques are designed specifically for waste minimization and waste classification control. For example, one method involves abrasive blasting with solid carbon dioxide (dry ice), which sublimates after use and can be exhausted through a HEPA filter, leaving no added material to the waste. Decontamination with high-pressure water has some similar advantages, but care must be taken to ensure that used decontamination solutions do not spread contamination.

### **8.3.3 Waste Treatments**

Available treatments for solid waste include compaction and incineration. In specific cases, there may be decontamination options available, as well.

Compaction, with pressures in the range of 40,000 to 60,000 psi, is most often used on paper, fabric, and plastic although it is effective on glass, sheet metal, and some other materials. With such ordinary materials, one commercial reactor has approached up to 800 pounds of waste per 55-gallon drum, although an average of 500 pounds per drum is considered very good. Compaction is done by drum compactor or box compactor. Compacting into a drum or a 4- by 4- by 8-ft box is normally a labor-intensive operation and often involves some risk of personnel exposure, even though the better compactors are equipped with HEPA-filtered ventilation systems. Supercompaction uses considerably higher pressures than compaction, normally 200,000 psi or greater. Supercompaction usually involves compacting filled waste drums into a box or overpack. Supercompaction has been successfully used on piping and other materials that are normally considered noncompactable.

It is really a choice of words whether incineration is considered a disposal technique or a volume-reduction technique. All carbon, oxygen (except for any that becomes bound in oxide ash), nitrogen, hydrogen, and sulfur present in the incinerator feed will be converted to gasses and disposed to the atmosphere. Plutonium and most metals will

remain as a solid material. As a volume-reduction technique, incineration is very successful, with volume-reduction factors up to 200:1 or greater achieved on some waste streams. There have been licensing delays for some incinerators, and often there are limitations brought about by air-quality restrictions. There is also the possibility that incinerator ash may be a mixed waste due to the concentration of other impurities such as heavy metals in the waste. If a facility has an incinerator, a quantity of the feed material can be incinerated to determine if the waste will have hazardous characteristics before the material is contaminated. In some cases, it is desirable to size-reduce or repackage in combustible packaging before incineration.

Decontamination is most successful when the material can be recycled for use in a nuclear facility since the need to prove releasability (cleanliness) is eliminated. Nevertheless, cleaning material for unrestricted release is also possible in some cases. It may also be possible to decontaminate an item enough to change its classification from TRU waste to LLW, thereby allowing immediate disposal of the item, while a relatively small quantity of decontamination waste is stored as TRU waste.

Electropolishing to remove the thinnest metal surface has been very effective and produces a relatively small waste volume, especially when one of the wetted sponge units is used rather than an emersion tank. Surface scabbling has been used in decontamination of concrete, and various abrasive blasting methods have also been effective. Strippable and self-stripping coatings may be used to decontaminate surfaces, even though the primary application of strippable coatings has been in preventing contamination of surfaces.

There are occasionally mixed strategies that work well. Used HEPA filters may be removed from their frame for compaction. Metal frames may be decontaminated and wood frames may be incinerated.

#### **8.3.4 Sampling and Monitoring**

Solid waste is monitored for several reasons: to determine if it can be released as sanitary (or hazardous) waste; to distinguish its classification as either LLW or TRU waste, depending on the concentration of transuranic isotopes; and to obtain defensible values

for documenting shipping and disposal quantities. See section 4.2.4.2 for guidance on release surveys.

### **8.3.5 Storage and Disposal**

Solid sanitary waste, hazardous waste, and LLW can normally be disposed of using existing procedures. Transuranic waste, HLW, and most mixed waste may have to be stored for a period of time awaiting approval of disposal facilities; they will have to be stored in a manner that prevents routine and accidental impact on the environment. They must be protected from unauthorized access, fire, flood, or water damage. Containers must be protected from corrosion or other deterioration and an accurate inventory of the material must be kept. Most facilities prefer to store such material in a form that they believe will be shippable.

Existing storage and packaging requirements for plutonium metal and oxide are addressed in DOE Order 460.1A (DOE, 1996b). The DOE's existing storage practices for plutonium and plutonium-containing materials and wastes were evaluated at a DOE Workshop in May 1993 [see Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a)]. The draft recommendations from this workshop for metals and oxides that are not in containment vessels with certified hermetic seals [per ANSI N14.5 (ANSI, 1987d)] are given in Table 8.3. The variety of plutonium-containing materials is illustrated by the inventory information for the Hanford Site contained in documents by Christensen et al. (1989) and Hoyt (1993).

## **8.4 LIQUID WASTE**

Liquid waste from plutonium facilities includes various aqueous waste streams such as cooling water, laundry waste, and floor-drain waste, and numerous organic and inorganic chemical wastes. The design criteria and operational controls to make these streams treatable and disposable, and the methods to treat them are beyond the scope of this document and are highly facility-specific. General considerations are given below.

#### **8.4.1 Design Objectives**

If a facility process requires the generation of plutonium-contaminated liquids, it is probably best to ensure that the mother liquid is demineralized water and that plutonium is the only contaminant added. In this case, the liquid can be filtered, demineralized, and recycled. Any other chemicals added to the water will complicate treatment, increase the volume of secondary waste, and diminish the opportunity for recycle. Organic contaminants such as oils, solvents, and detergents will likely foul the ion exchange resin, greatly increasing resin volume.

A pure organic solvent has many of the advantages of demineralized water, especially if it does not chemically degrade or evaporate under the conditions of use. (Solvents are not usually amenable to purification by ion exchange; however, filtration, extraction into aqueous solutions, and distillation are possible.) Unfortunately, most organic solvents are classified as hazardous materials and any material that comes in contact with them is likely to be a hazardous (or mixed) waste when it is disposed of. If the solvent is combustible and the facility includes an approved incinerator of sufficient capacity to handle the secondary waste, then the organic solvents are highly desirable.

While such guidance may be helpful in facility design, there will be waste streams that do not conform to either of the situations above. Most decontamination wastes, laundry wastes, and floor-drain wastes are examples. In decontamination, it is important that the process is selected with provisions to manage the waste. In many cases, the nature of the facility determines that the waste will be a mixed waste. In these cases, minimizing the volume is most important. For example, if a plutonium-contaminated surface has been painted with a lead-based paint, the decontamination waste will be mixed waste unless it is further treated to ensure that the lead is not in a leachable form. In this example, removing the paint by dry ice blasting, high-pressure water blasting, heat, or a similar method would be preferable to sand blasting in which the sand would be added to form an additional mixed waste that could require storage for many years.

Laundry wastes are a special problem because radioactive contamination, body oils, and odors must be removed from protective clothing. For a time, dry cleaning was extremely popular, because the solvents were easily redistilled and recycled. However, because the

solvents were usually chlorofluorocarbons and because the small volume of waste generated was mixed waste, this method is now rarely used. Incineration of disposable protective clothing is an outstanding choice if an incinerator of sufficient capacity is available, but this is rarely the case. Water washing is often the method of choice. In a few cases, plutonium in the waste stream is removed adequately by filtration and the effluent can be disposed to a sanitary sewer or to the environment under a National Pollutant Discharge Elimination System (NPDES) permit. It is important to select a detergent for water washing that does not foul or plug the filter and that has a minimal impact on ion exchange resins if they must be used. Many household laundry detergents have fillers such as wood fiber to give them greater bulk. These should never be used because the fiber has no beneficial use and will end up as solid waste. As a general rule, extensive testing on clean material should be done to optimize disposal of laundry waste.

Floor-drain wastes are much more of a problem in some facilities than in others. In some facilities, there is a culture that says, “if you don't know what to do with it, pour it down the floor drain.” Such practices can lead to a mixture of water, detergent, oil, antifreeze, and other substances that clog filters and foul ion exchange resins. In the worst cases, solidification with Portland cement is the only alternative, and this increases an already large volume. The use of catch basins under chemical and lubricating systems and extensive training of personnel minimize the probability of such occurrences. Oil skimmers on floor-drain collection tanks are sometimes advisable, as well.

#### **8.4.2 Operational Controls**

Once the facility is properly designed, training of personnel is the primary operational control against generating excessive volumes of waste or against generating waste with contaminants that interfere with treatment or change the classification.

Some facilities have used color codes to prevent materials from entering an area where they will adversely affect waste management. For example, certain electronic contact cleaners may be banned from some radiologically contaminated plant areas because they would generate mixed waste. The procurement organization might code all such materials red and certain areas would be posted to indicate that the materials were not allowed.

Whatever the system, it is important that each employee be trained to effectively use the system and that well-intentioned housekeeping efforts do not result in excessive waste volumes.

### **8.4.3 Waste Treatments**

The primary treatments for aqueous waste are

- pH adjustment
- precipitation
- liquid-solid separation such as flocculation and filtration
- ion exchange
- distillation
- purification by reverse osmosis
- solidification.

The primary treatments for organic solvents are:

- Solvent extraction
- filtration
- incineration.

Virtually all of these processes (except pH adjustment) are likely to result in secondary waste that requires treatment and/or disposal. In all cases, recycling of the primary solution is desirable because it reduces monitoring cost and waste-disposal liability and cost. A brief description of the use of each of these treatment methods is given below. Extensive design and engineering should be done before any method is selected in order to ensure meeting design objectives.

#### **8.4.3.1 pH Adjustment**

This treatment is used on aqueous systems to meet discharge limitations or to make the solution amenable to other treatment. A mineral acid, such as sulfuric, hydrochloric, or nitric, is normally used to lower the pH. A base, such as



sodium, potassium hydroxide, or occasionally ammonia, is used to raise the pH. The solubility of some contaminants will be affected by the pH of the solution. For example, an acidic solution containing iron may show a copious precipitate of ferric hydroxide upon the addition of a base.

#### **8.4.3.2 Precipitation and Co-precipitation**

Precipitation and co-precipitation are used to decrease the solubility of some compounds. Precipitation involves making the contaminant into an insoluble material by the adjustment of pH or the addition of a chemical. For example, nickel may be rendered insoluble by the addition of sodium dimethylglyoxime. Co-precipitation is similar but is used when the contaminant is not present in sufficient quantity to form a filterable solid but will incorporate into another precipitate as it forms or will adhere to the surface of another precipitate. In some waste treatment processes, a stable isotope of the radioactive contaminant is added to co-precipitate the radioactive material that is not present in sufficient quantity to form a precipitate on its own. Precipitation is always followed by some liquid/solid separation technique.

#### **8.4.3.3 Liquid-Solid Separation Techniques**

Treatments such as flocculation and filtration are used to remove solid and colloidal contaminants either directly from the waste stream or following a precipitation or co-precipitation process. Centrifugation or settling are sometimes used to remove gross quantities of solids preceding some filtration processes. These processes separate the waste into a concentrated and dilute waste stream, both of which will probably require further treatment. The bulk liquid fraction may be subject to filtration before recycling or disposal. The fraction with the high concentration of solids may be subject to evaporation, or drum or filter-press filtration to remove excess water, or it may be solidified as discussed below.

Where the contaminant is present as a colloid or extremely fine particulate, co-precipitation or flocculation may be required before settling, centrifugation, or

filtration. Flocculation involves the addition of an extremely small quantity of a long chain molecule that has the appropriate electrostatic affinity for the contaminant present. The flocculent molecules gather the contaminant into rather large particles that are amenable to settling and filtration. The flocculent and dosage (addition ratio) are usually selected by trial and error. Flocculents do not add appreciably to the waste volume and usually do not add a contaminant that results in a mixed waste. Residual flocculent may, however, foul ion exchange resins or reverse osmosis membranes, so it is important that the quantity added be closely controlled.

#### **8.4.3.4 Ion Exchange**

Ion exchange is one of the most useful waste treatment techniques. Aqueous wastes that are free of oil and other organics and contain only very minimal quantities of solids may be subject to ion exchange on cation resin, anion resin, or specialty resins, either alone or in combination. If the contaminant is present as a cation, such as sodium, ammonia, or calcium, a cation resin can be used to replace the cation in solution. The cation from the resin will go into the solution to replace the contaminant cation. If the water stream is being recycled, the cation resin will probably be in the hydrogen form so that only hydrogen ions will enter the solution. If a hydrogen form of cation resin is used by itself, the water solution will likely become more acidic (lower pH). If an anion resin is used, anions in solution will be replaced with anions from the resin. Although resin may be in a chloride or other form, the hydroxyl form of the resin is often used so that anions are replaced with hydroxyl anions (-OH). If only a hydroxyl anion resin is used, the solution will drop in pH, becoming more basic. If both a hydrogen form of cation resin and a hydroxyl form of anion resin are used, the ions they add combine to form water, so both resins are used on demineralized water systems that are recycled. One disadvantage of most ion-exchange resins for waste treatment is the fact that they remove all ionic contaminants, not just the radioactive ones, and so are exhausted earlier than they might be. Selective resins are available for a few materials, most notably cesium, but are not available for plutonium.

In some applications, radionuclides pass through both cation and anion resin beds. This is assumed to happen because they are not present in an ionic form. They are either colloidal or are present in a molecule or complex that is neutral. In these cases, pretreatment or multiple treatment steps may be required.

Unfortunately, plutonium may be present as a cation, anion, neutral chemical complex, or colloid. Testing is almost always required to optimize plutonium removal. One additional limitation in the use of most ion exchange media for plutonium and other alpha-emitting radionuclides is that the radiation degrades the resin over time. Organic ion exchange media loaded with large quantities of plutonium may emit hydrogen and may become unstable when exposed to oxidizing materials such as nitric acid.

In some applications, ion exchange resins are "recharged" by the addition of large quantities of a particular ion (e.g., hydrochloric acid may be used to reconvert spent cation resin to the hydrogen form). In nuclear applications, this is rarely feasible because of the need to dispose of the recharge solution and because of the large quantity of rinse water used to remove the excess recharge solution from the resin.

#### **8.4.3.5 Distillation**

Distillation (including vacuum distillation) is at least conceptually simple. It removes all but volatile contaminants. In practice, some contaminants will cause foaming, and evaporator maintenance is often a problem. If laundry waste or other waste-containing detergents are to be evaporated, it may be necessary to add an antifoaming compound. Although these are sometimes effective, they often degrade with heat faster than the detergents or other compounds causing the foaming. Few evaporators take the product to dryness, as this often creates a scale build-up. If the evaporator bottoms are removed as a solution, they must be solidified, usually with some increase in volume.

#### **8.4.3.6 Purification by Reverse Osmosis**

This process is highly effective on relatively pure water streams. The water is passed through a semipermeable membrane by mechanical pressure, leaving contaminants behind. The result is generally 80% to 99% of the influent water released as pure water, with the remainder containing all of the contaminants. Reverse osmosis has the advantage over ion exchange in that it will remove non-ionic contaminants although these often shorten the life of the membrane. It is much more energy-efficient than distillation and requires much less equipment for the same volume of water treated. It is sometimes used as a "polishing" technique to further treat relatively clean water.

#### **8.4.3.7 Solidification**

Solidification is often a last-resort treatment because, while the other treatments described reduce the volume of solid waste requiring disposal, solidification increases it. Nevertheless, it is useful for some waste. Portland cement is the most common solidification medium for water solutions, aqueous suspensions, and resins. However, there are other proprietary materials, including some especially for oils and other organic compounds.

#### **8.4.3.8 Solvent Extraction**

Solvent extraction is used exclusively with organic solvents and involves mixing the solvent with an immiscible aqueous solution in which the contaminant is soluble. In this way, the contaminant is transferred to the aqueous solution for further treatment. (Solvent extraction may also be used in the other mode, in which the contaminant is transferred to the organic solvent solution, but this has fewer applications in waste management.) The organic solution is usually recycled.

#### **8.4.3.9 Incineration**

Incineration is an ideal waste-management technique for combustible solvents and other liquids that do not yield toxic or hazardous combustion products. The volume reduction from feed material to ash is usually outstanding. Incinerators are usually equipped with wet scrubbers, demisters, and filters to ensure that the effluent released to the environment is acceptable and ALARA. These features create secondary waste that must be dealt with, but the disposal efficiency usually makes them well worthwhile.

#### **8.4.4 Sampling and Monitoring**

Sampling and monitoring of liquid waste streams are usually straightforward. Bulk liquid in tanks must often be mixed, usually with a recirculating pump, before dip sampling to ensure a representative sample. Liquid effluent streams are often sampled with a flow-proportional sampler. For on-line monitoring, a small ion exchange column is used to concentrate ionic contaminants, and a detector is placed on the column for gamma analysis.

#### **8.4.5 Storage and Disposal**

Sanitary liquids and those meeting disposal criteria may be released to the environment or to sanitary waste treatment systems (sewerage systems). Hazardous liquid waste may be shipped, with excess absorbent material in compliance with 40 CFR, to a licensed treatment facility. Small quantities of radioactive-contaminated liquids, such as samples, may be shipped in a similar way, but most liquid waste must be solidified prior to shipment or disposal. It is preferable to store only solid waste, as well. The recommendations of Table 8.3 are applicable to the storage of plutonium-containing liquids as well as solids. In particular, where long-term storage of plutonium solutions may occur, even within glove boxes, it is advisable to avoid plastic containers unless one can be certain that the alpha radiation will not have degraded the container.

**Table 8.3.** Interim Recommendations for Storage of Plutonium Metal and Plutonium Oxide at Department of Energy Facilities

<p>The following recommendations are made to improve current plutonium storage safety practices. Until new equipment and facilities become available to package plutonium based upon long-term standards, these recommendations are applicable to plutonium metal or plutonium oxide stored outside of glove-box lines in containers that do not have certified hermetic seals (i.e., per ANSI N14.5 (ANSI, 1987d)).</p>	
1.	Plutonium solutions, metal turnings, or particles with specific surface areas greater than 1 cm <sup>2</sup> /g should not be stored outside of glove boxes.
2.	All packages containing plutonium metal should be taped, re-taped, and placed in plastic bags prior to handling.
3.	Inspections should incorporate use of adequate personnel protection. Inspection practices should be codified in surveillance plans. These plans should reflect current facility operating status. There must be personnel radiological surveillance during all handling operations. Personnel protection during operations should include protective clothing and gloves and, if necessary, respiratory protection.
4.	Inspection of containers should be integrated with audits for materials control and accountability (MC&A) to minimize container-handling and attendant radiation exposure to ALARA levels.
5.	Containers should be inspected for abnormalities (e.g., mass change, container deformation, or discoloration) using visual inspection, weighing, or video surveillance where such capability exists. Findings should be recorded for safety and MC&A evaluations. Visual inspections should be made at intervals of 1 week and 1 month after the material's initial containment and annually thereafter.
6.	Packages containing more than 0.5 kg of plutonium metal should undergo an annual surveillance in which the total mass of the package is determined to an accuracy of $\pm 0.5$ g and compared with the preceding year's mass and with the initial (reference) mass at the time of packaging. A storage package should be evaluated (e.g., opened and inspected, radiographed) if any of the following conditions are evident:
	<ul style="list-style-type: none"> <li>a. The outer storage vessel is bulged or distorted.</li> <li>b. Hydride-catalyzed oxidation is suspected. Such reaction is indicated by a mass increase in either of two circumstances:</li> </ul>

**Table 8.3 (cont'd)**

	<p>i) For packages whose masses continue to increase since initial packaging or for which historical mass data are unavailable (see item 6 above), a mass increase greater than 15 g per kilogram of plutonium over a one-year period indicates a hydride-catalyzed oxidation reaction.<sup>(a)</sup></p> <p>ii) For a package whose mass has remained constant over a period of several years (less than <math>\pm 0.5</math> g change) from its reference value, then undergoes an annual mass increase of more than 2 g per kilogram of plutonium, hydride-catalyzed reaction is indicated. Such a package is particularly suspect. The indications are that a previously sealed container may now be breached and that the continuing reaction may lead to rapid containment failure within 12 to 24 months</p>
	<p>c. The measured package mass, relative to the reference mass, corresponds to the mass that indicates formation of oxide with a volume exceeding 10% of the free volume of the inner vessel. Each 1-g increase in mass corresponds to formation of 1.5 cm<sup>3</sup> of oxide with a density of 50% of the theoretical value of 11.46 g/cm<sup>3</sup>.</p>
7.	<p>Inspected containers exhibiting abnormalities (e.g., external contamination, bulging, discoloration, or other anomalies) should be repackaged in accordance with well-defined procedures (see items 3 and 4 above). Handling such containers outside of a glove box or conveyor confinement requires respiratory protection until the package is placed in an overpack container (e.g., taped metal can or sealed plastic bag) before further handling and transport.</p>
8.	<p>As an interim measure, material that is repackaged may be placed in a food pack can or slip-fit (Vollrath) container with a secured lid. If possible, metal should be repackaged in a configuration containing at least one gas-tight seal. No plastic material should be in direct contact with plutonium metal or oxide, and use of plastic in outer layers of packaging should be minimized.</p>
9.	<p>When packaging metal, hazardous or pyrophoric material such as plutonium hydride should be removed. However, it is not necessary to remove protective oxide film. Metal should be packaged in as dry and inert an environment as possible to minimize corrosion (&lt;100 ppm H<sub>2</sub>O).</p>

**Table 8.3 (cont'd)**

10.	Impure oxide from sources other than metal should be thermally stabilized at $1000 \pm 100^\circ\text{C}$ for at least an hour, or placed in a combination of a slightly lower temperature ( $\sim 850^\circ\text{C}$ ) for longer heating time to result in the lowest loss on ignition (LOI) practicable with existing equipment. This ensures complete conversion of substoichiometric material and aids small-particle coalescence, which diminishes dispersal risk.
11.	Because plutonium oxide has greater potential for dispersion in severe accidents, it should have priority over metal for storage in structurally robust vaults. Metal should be characterized to ensure that it has not converted to oxide while in storage. Stored plutonium will have an increasing radiation level because of the build-up of $^{241}\text{Am}$ . Therefore, characterization of metal should be done as soon as possible and should make full use of small-sample statistical methods to minimize worker exposure. The results of characterization should be integrated with a site's surveillance plan, as well.
12.	Quality assurance measures, labeling, and material characterization are essential. Material and storage packaging specifics should be thoroughly documented.

- (a) A higher oxidation rate may occur if the contained metal exhibits a high surface-area configuration, such as sheet or foil. The maximum annual increase for normal (uncatalyzed) oxidation of a given metal geometry can be calculated using a reaction rate of  $3 \times 10^{-7}$  g oxygen/cm<sup>2</sup>-minute measured for alpha-phase plutonium under moist conditions at  $50^\circ\text{C}$ .



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## **9.0 EMERGENCY MANAGEMENT**

It is DOE policy that all DOE facilities and activities be prepared to respond to operational emergencies in a way that minimizes consequences to workers, the public and the environment. Formal emergency management programs are the final element of DOE's defense-in-depth against adverse consequences resulting from its operations.

### **9.1 Emergency Management In DOE**

DOE Order 151.1 (DOE, 1995a) requires DOE elements and contractors to plan and prepare for the management of emergencies. The following discussion of emergency management principles, requirements and guidance is generally applicable to DOE plutonium facilities. Specific facility requirements are in accordance with the individual facility DOE contract.

#### **9.1.1 Key Emergency Management Principles**

DOE emergency management policy and direction is based on four key principles: planning and preparedness commensurate with hazards; integrated planning for health, safety and environmental emergencies; classification of and graded response to emergencies, and; multiple levels (tiers) of emergency management responsibility.

NOTE ON TERMINOLOGY: Within the EMS, “planning” includes the development of emergency plans and procedures and the identification of personnel and resources necessary to provide an effective response. “Preparedness” is the procurement and maintenance of resources, training of personnel, and exercising of the plans, procedures, personnel and resources. “Response” is the implementation of the plans during an emergency to mitigate consequences and to effect recovery.

- a) **Planning and Preparedness Commensurate with Hazards.** Because of the wide range of activities and operations under DOE's authority, standards and criteria suited to one type of facility or hazard may be inappropriate for another. To deal with this diversity, while assuring an adequate overall state of preparedness, DOE Orders specify standards for the structure and features of

emergency management plans and require that the details of each feature be tailored to the unique hazards of the specific facility. This approach ensures a more complete and quantitative understanding of the hazards while providing for focused and cost-effective emergency planning and preparedness.

- b) **Integrated Planning for Health, Safety and Environmental Emergencies.** A wide variety of different types of operational emergencies can occur at DOE operations. Some may involve loss of control over radioactive or other hazardous materials unique to DOE operations, while others may involve security, transportation activities, natural phenomena impacts, environmental damage, or worker safety and health concerns. Planning, preparedness and response requirements for some types of emergency conditions are specified by other agencies having authority over DOE facilities and activities. For example, Federal regulations on occupational safety, environmental protection and hazardous waste operations have consequent “emergency planning” requirements. Rather than meet these requirements piecemeal through separate programs, DOE has combined, under the EMS, all planning and preparedness activities for emergency events having health, safety or environmental significance.
- c) **Classification of Emergencies and Graded Response.** Operational emergencies involving hazardous materials are grouped into one of three classes according to magnitude or severity. Classification of events is intended to promote more timely and effective response by triggering planned response actions generally appropriate to all events of a given class. This principle, termed “graded response”, is embodied in DOE Order requirements and is important to the effective management of response resources.
- d) **Tiers of Emergency Management Responsibility.** Within the EMS, responsibility for emergency management extends from the individual facility level to the cognizant DOE Field Element, and culminates at the cognizant Headquarters Program Office. The responsibilities vested at each level of the hierarchy are specified in DOE Order 151.1 (DOE, 1995a). The responsibility and authority for recognizing, classifying, and mitigating emergencies always

rests with the facility staff. The head of the cognizant Field Element oversees the response of contractors and supports the response with communications, notifications, logistics, and coordination with other DOE elements. The DOE Headquarters (HQ) Emergency Operations Center (EOC) receives, coordinates, and disseminates emergency information to HQ elements, the cognizant Program Office, Congressional offices, the White House, and other Federal Agencies.

### **9.1.2 Requirements Pertaining to All DOE Operations**

DOE Order 151.1 (DOE, 1995a) identifies standard program elements that comprise each DOE facility emergency management program. The elements form a standard framework, with the details of each program element varying according to the nature and magnitude of the facility hazards and other factors. The Orders require that a facility-specific hazards assessment be conducted and the results used as the technical basis for the program element content. Using the results of an objective, quantitative, and rigorous hazards assessment as a basis, each program is configured to the specific hazards and response needs of the facility.

Detailed guidance on the implementation of the Order requirements is being published by the DOE Office of Emergency Management (DOE, 1997). The Emergency Management Guides (EMGs) have been drafted, put through final concurrence, and are awaiting publication. The EMGs specify acceptable methods of meeting the Order requirements. Individual guides have been published for the hazards assessment process and for program elements.

## **9.2 Specific Guidance on Emergency Management For Plutonium Facilities**

This section provides technical guidance that is specifically applicable to the development and implementation of emergency management programs for plutonium facilities. It is intended to supplement, not replace, the more general recommendations provided in the EMGs.

### 9.2.1 Hazards Assessment

The emergency management hazards assessment for a facility that stores or processes plutonium or its compounds should follow the basic assessment process outlined in the DOE Emergency Management Guide. Guidance for Hazards Assessment (DOE, 1992a). The Emergency Management Hazard Assessment (EMHA) should be based upon the facility specific Safety Analysis Report (SAR), which develops facility and operations hazards. Unique properties and characteristics of plutonium and its compounds may need to be considered at certain steps in the hazards assessment process.

- a) **Description of Facility and Operations.** The properties of the hazardous material do not significantly affect the manner in which this step of the hazards assessment is performed, except to the extent that plutonium safety considerations may mandate more detailed descriptions of certain facility physical or operational features.
- b) **Identifying and Screening the Hazards.** The objective of this step is to identify hazards that are significant enough to warrant consideration in a facility's emergency management program. It is recommended that screening thresholds (or quantities) be selected for each hazardous material. This screening threshold value is then compared to the inventories of the material at risk of being released from a single event. If a particular inventory of material is less than the screening threshold value, the consequences of its release are presumed to be minimal. The potential release of that inventory need not be analyzed further.

The screening threshold value should be based on the dominant hazardous property of a material. For all plutonium isotopes and all its chemical forms, radiotoxicity is the property of most concern.

Several possible sources of screening threshold values are suggested by the EMG. For radioactive materials, the primary source of screening threshold values is 10 CFR 30.72 Schedule C, *Quantities of Radioactive Materials Requiring Consideration of the Need for an Emergency Plan for Responding to a Release* (CFR, 1992a). For plutonium, the most

applicable number from that source is the 2 Curie value for "all other alpha emitters". However, facilities may select lower screening values based on the properties of the material. A screening threshold can be determined by modeling a unit release of the material to the atmosphere at ground level and determining the consequences at some reference distance under conservative dispersion conditions. The screening threshold value is typically a quantity which, if released, would produce consequences on the order of one-tenth the threshold for protective action at the facility boundary.

The quantity of plutonium needed to achieve criticality, even under optimum conditions of moderation and reflection, is sufficiently large that the radiotoxicity of the plutonium itself will always serve as the basis for determining whether a given inventory does or does not need to be analyzed.

- c) **Characterizing the Hazards.** The objective of this step is to describe the hazardous materials in sufficient detail to allow accurate modeling of releases and calculation of consequences.

The following properties of plutonium and its compounds influence the release potential and consequences.

- Chemical and physical form. The chemical toxicity of plutonium and its compounds is of much less concern than the radiotoxicity of the plutonium. However, the chemical and physical form may strongly influence the release potential. Plutonium metal oxidizes readily in humid air at elevated temperatures to form loosely-attached oxide particles, a source of readily dispersible airborne and surface contamination. Plutonium metal fines and turnings can ignite spontaneously in the presence of air, creating aerosol-size oxide particles and providing energy to disperse them. Also, some plutonium compounds may ignite violently on contact with air, water or hydrocarbons (Benedict, et al., 1981).

- Solubility. The committed effective dose equivalent (CEDE) per unit activity inhaled is about three times greater for plutonium of solubility class W than for class Y. No plutonium compounds of solubility class D are generally recognized.
  - Particle size. Particle size distribution has a large effect on the radiotoxicity of inhaled materials. Larger particles tend to be cleared rapidly from the upper respiratory regions and swallowed, thereby delivering little radiation dose to the lung tissues. Because plutonium is poorly absorbed in the gut, very little dose is attributed to the larger particles that are cleared from the body by this process. Small particles are deposited deeper in the lung and are cleared very slowly, producing a much larger dose per unit activity inhaled. Extremely small particles tend to be exhaled and not deposited.
  - Isotopic mixture. Characterization of the isotopic mixture is important to the accuracy of both dose calculations and contamination measurements. When the inventory or quantity released is expressed as the total activity (Ci or Bq) of a mixture of isotopes, the total often includes the  $^{241}\text{Pu}$  activity. Because  $^{241}\text{Pu}$  decays almost exclusively by beta emission, it contributes little to the internal dose from a mixture of Pu isotopes. Also, the fraction of  $^{241}\text{Am}$  (from decay of  $^{241}\text{Pu}$ ) in plutonium can vary greatly, depending on the degree of irradiation and the time since the plutonium was chemically separated from the reactor fuel. Characterization of contamination from a plutonium mixture is often done by detecting the low-energy photons emitted by  $^{241}\text{Am}$ , which requires knowledge of the activity of  $^{241}\text{Am}$  compared to the other isotopes in the mixture.
- d) **Developing Event Scenarios.** The properties of the hazardous material do not significantly affect the manner in which this step of the hazards assessment is performed.

- e) **Estimating Potential Event Consequences.** For the scenarios developed in the previous step, this step determines the area potentially affected, the need for protective actions, and the time available to take those actions. The way these consequences are determined depend on properties of the hazardous material.

For plutonium and its compounds, inhalation during plume passage is the most important exposure process in the early phase of an emergency.

After passage of a plume, exposure to material deposited on the ground will dominate. Therefore, the following features should be considered when selecting and applying calculational models:

- Inhalation pathway dose. For any realistic mixture of plutonium isotopes, the great majority of the dose will be by the inhalation pathway. Therefore, the model selected to estimate consequences of an atmospheric plutonium release must be able to calculate the total effective dose equivalent (TEDE) to an individual exposed by inhalation.
- Plume depletion during transport. As it is transported downwind, an aerosol plume will be depleted by gravitational settling of particles. Because of the high density of plutonium and its compounds, this depletion effect can be very significant in reducing the dose. Therefore, a consequence model that accounts for plume depletion by gravitational settling should be used. When analyzing consequences of any postulated accidental criticality, any model selected should account for the decay during transport of short-lived fission product gases.
- Ground deposition. Following passage of a plume, the amount of plutonium deposited on the ground will determine whether long-term intervention to minimize the dose to the resident population will be required. The consequence model selected should calculate ground deposition to support protective action planning.

### 9.2.2 Program Elements



Properties and characteristics of plutonium and its compounds must be considered in formulating the emergency management program elements. Following are specific program element considerations related to the hazardous properties of plutonium.

- a) **Emergency Response Organization.** The primary influence of plutonium's hazardous properties on the Emergency Response Organization (ERO) is in the staffing of the consequence assessment component. As will be discussed in e) below, staff should be assigned to the ERO who are knowledgeable of and able to quantitatively evaluate the radiological aspects of the hazard.
- b) **Offsite Response Interfaces.** The specific properties of the hazardous material do not significantly affect the content of this program element.
- c) **Operational Emergency Event Classes.** As with all hazardous materials, classification of emergencies for plutonium facilities should be based on the predicted consequences at specific receptor locations, as compared with numerical criteria for taking protective action (total effective dose equivalent). The classification of the postulated event or condition should be determined during the hazards assessment process and the observable features and indications identified as Emergency Action Levels (EALs) for that event/condition.
- d) **Notification.** The specific properties of the hazardous material do not significantly affect the content of this program element.
- e) **Consequence Assessment.** As discussed in section 9.3.1 c) and e), models and calculational methods used for consequence assessment should be appropriate to the physical, chemical, and radiological properties of the hazards. Models used to calculate and project the radiological consequences of a release of plutonium should be the same ones used in the hazards assessment process. If the same models are not used, the differences between outputs should be characterized and documented to avoid the potential for confusion and indecision during response to an actual emergency. The facility SAR consequence assessment of identified

hazards should be used as a starting point for detailed EMHA plutonium consequence assessment.

Environmental monitoring capability for assessing consequences of a plutonium release should conform to several general principles.

- Procedures for measurement of airborne plutonium should provide for timely analysis and reporting of results in units that correspond to decision criteria. Decision points based on initial alpha screening measurements with field instruments should account for the expected levels of radon progeny collected on the air sample media. Alternatively, portable survey instruments capable of performing alpha spectroscopy measurements can be used to provide rapid isotopic analysis of plutonium collected on sample media.
- Measurement of plutonium deposition should be planned and proceduralized to yield results that correspond to those needed by the predictive models used for emergency response. The correlation between direct or indirect radioactivity measurements (in units of activity) and measurement methods that give mass or concentration of plutonium in a sample should be established for standard sample sizes, collection efficiencies, and the expected isotopic mixture(s) of material that might be released. Information on expected isotopic mixture should be available for converting the results of measurements made with photon-sensitive instruments, such as the Fiddler and Violinist, into plutonium activity per unit area.
- If the potential exists for release of plutonium in conjunction with materials of high chemical toxicity, it is generally not practical to plan on use of survey teams to quantify concentrations in a plume. The high risk to survey personnel, the protective equipment necessary to minimize that risk, the time needed to prepare and deploy a team for such a survey and the limited value of the information that could be gained all weigh

against this approach to assessing the consequence of a highly toxic release.

- Continuous environmental air samples are taken around the perimeter of some plutonium facilities for environmental reporting purposes. Consequence assessment procedures should provide for the rapid retrieval and analysis of sample media from any fixed samplers that may be operating in an area affected by a plutonium release. The procedures should specify the type of measurements to be done on those sample media, including any instrument settings, conversion factors, or adjustments needed to produce useful results in the shortest time possible.

- f) **Protective Actions.** The Protective Action Guides (PAGs) published by the U.S. Environmental Protection Agency (EPA, 1991) have been adopted by DOE as its basic protective action criteria for planning and response. The evacuation PAG for the early phase of a radiological incident is 1 rem (TEDE) under normal conditions and up to 5 rem under conditions where evacuation might pose excessive hazards to the population (adverse weather, etc.).

Because the dose from intake of long-lived radionuclides like plutonium is delivered over a period of many years, the dose increment actually received in any given year is a small fraction of the total dose commitment. The Emergency Management Guide for the Implementation of DOE Order 151.1 (DOE, 1997) authorizes use of the 5 rem protective action threshold for planning and hazards assessment purposes when a large fraction of the dose from a radionuclide mixture will be delivered over a period of years following the exposure.

- g) **Medical Support.** If the potential exists for large intakes of plutonium, the emergency management program should include specific planning for the quantification of exposure, diagnosis of health effects, and treatment. Medical facilities providing emergency medical support should be provided with references relating to plutonium toxicity and treatment protocols. Criteria for

implementing treatments such as surgical excision of contaminated tissue, lung lavage, or use of chelating agents should be discussed with the medical staff and sources of real-time advice and assistance should be identified.

- h) **Recovery and Reentry.** The specific properties of the hazardous material do not significantly affect the content of this program element.
- I) **Public Information.** The specific properties of the hazardous material do not significantly affect the content of this program element.
- j) **Emergency Facilities and Equipment.** Except for instruments and analysis methods used in consequence assessment, little by way of specialized facilities and equipment will be required to meet the emergency management program needs of plutonium facilities. Equipment and analytical techniques for detection and measurement of plutonium in environmental sample media should have sufficient sensitivity to measure levels at or below those corresponding to decision criteria. Whereas larger sample sizes, chemical processing, or longer counting times may be used to reduce the limit of detection for routine environmental surveillance, time constraints may dictate that more sensitive techniques be available to meet the information needs of emergency response.
- k) **Training, Drills and Exercises, Program Administration.** The specific properties of the hazardous material do not significantly affect the content of these program elements.

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## **10.0 DECONTAMINATION AND DECOMMISSIONING**

At the end of the useful life of a facility, activities are undertaken to restore the facility to noncontaminated status and permit its unrestricted use. These activities are typically termed decontamination and decommissioning (D&D).

Although plutonium facilities are no longer useful and operational activities are no longer conducted, measures must be continued to control the residual radioactivity. The decision may be made to undertake a D&D program to minimize or eliminate long-term institutional control. This may be done in a variety of ways, most of which may be termed D&D. The exception is converting the facility to some other nuclear use. With the elimination of the DOE weapons production mission, more plutonium-contaminated facilities will require D&D in the near future.

This chapter provides guidance on establishing and implementing an effective D&D program. Major topic areas include regulations and standards, design features, D&D program, D&D techniques, and D&D experience. This chapter concentrates on the radiation-protection aspects of D&D at plutonium-contaminated DOE facilities.

## **10.1 REGULATIONS AND STANDARDS**

The standards that apply to the decommissioning of a plutonium-contaminated facility include virtually all of those that were applicable during facility operations, (e.g., 10 CFR 835, DOE P 450-1 and DOE Order 440.1) plus some additional ones such as 10 CFR 835.1002(d). The occupational safety and radiation dose limits, safety management requirements, radioactive and hazardous chemical disposal regulations, and transportation requirements are unaffected by the activity to which they apply.

No single DOE regulation covers all D&D requirements due to the wide variety of issues encompassed by D&D. These issues include project management, environmental surveillance, health and safety of workers and the public, engineering design, characterization survey techniques, D&D techniques, waste management, and waste transport. The primary DOE Orders pertaining to D&D activities are DOE Order 5820.2A, Radioactive Waste Management (DOE, 1988b); DOE Order 4700.1, Project Management System (DOE, 1992e); DOE Order 5400.5,

Radiation Protection of the Public and Environment (DOE, 1990b); DOE Order 6430.1A, General Design Criteria (DOE, 1989b); DOE Order 231.1 Environment Safety and Health Reporting (DOE, 1995b); DOE Order 420.1 Facility Safety (DOE, 1995c). The DOE operations offices may have implementation procedures corresponding to these Orders that contractors will also need to comply with.

Section 5 of DOE Order 5820.2A, Radioactive Waste Management (DOE, 1988b), provides requirements important to decommissioning of radioactively contaminated facilities. It requires that DOE organizations develop and document their programs to provide for the surveillance, maintenance, and decommissioning of contaminated facilities. Requirements are divided into the following categories: general, facility design, post-operational activities, decommissioning project activities, and quality assurance. These categories are discussed below in Section 10.3.

DOE Order 4700.1, Program Management System (DOE, 1992e), provides the requirements to ensure a disciplined, systematic, and coordinated approach to project management. All projects, including D&D projects, should have clearly defined goals and objectives that support program requirements. Specific objectives include 1) promoting project execution that meets technical, schedule, and cost objectives, 2) meeting all applicable environmental, health and safety, and quality assurance requirements, and 3) avoiding a commitment of major resources before project definition. Good program management techniques should consider D&D costs as part of the life-cycle cost and select a tentative D&D method during the facility design phase.

Section V of DOE Order 5400.5, Radiation Protection of the Public and Environment (DOE, 1990b), provides radiological protection requirements and guidelines for cleanup of residual radioactive material and management of the resulting wastes and residues and release of property. This DOE Order establishes a basic public dose limit for exposure to residual radioactive material (in addition to naturally occurring “background” exposures) of a 100-mrem (1-mSv) effective dose equivalent in a year. A more detailed discussion is presented below in Section 10.1.3.

DOE Order 6430.1A, General Design Criteria (DOE, 1989b), provides design principles that shall be considered when designing radioactive material facilities to facilitate D&D of these facilities. A more detailed discussion of these design principles is found below in Section 10.2.

DOE O 420.1, Facility Safety (DOE, 1995c), establishes facility safety requirements related to: nuclear safety design, criticality safety, fire protection and natural phenomena hazards mitigation.

DOE O 231.1 Environment, Safety and Health Reporting (DOE, 1995b), ensures collection and reporting of information on environment, safety and health that is required by law or regulation to be collected, or that is essential for evaluating DOE operations and identifying opportunities for improvement needed for planning purposes within the DOE.

### **10.1.1 Other Regulations**

The D&D of most plutonium-contaminated facilities will involve cleanup of a combination of radioactive wastes, hazardous wastes, and mixed wastes. Some other Federal regulations not already discussed that are applicable to the cleanup and disposal of these wastes are summarized in this section along with the DOE guidance on implementation. This is not an all inclusive list. It is the facility responsibility to identify applicable requirements and ensure compliance.

- National Environmental Policy Act (NEPA) (USC, 1970) and 40 CFR 1500 (CEQ, 1992) - This act established a national policy to ensure that environmental factors are considered in any Federal agency's planning and decisionmaking. DOE O 451.1, National Environmental Policy Act Compliance Program (DOE, 1995d), defines DOE responsibilities and procedures to implement NEPA. The decommissioning of a DOE plutonium facility will require a determination of whether or not the action is a "major or significant government action adversely affecting the environment" in accordance with NEPA. If it qualifies as such an action, an environmental assessment (EA) or environmental impact statement (EIS) will be required. An EA or EIS will need to discuss the amount of material that will remain onsite and its effect, in addition to addressing the alternatives. The alternatives will include retaining radioactive material onsite under DOE control, cleaning the site to a level that would be acceptable for unrestricted release, and the null or no-action alternative of "walking away" from the site. If the action does not require an EA or EIS, either because the possible adverse



effects are insignificant or because decommissioning was adequately addressed in a preoperational or other EA or EIS, then the decommissioning can proceed in accordance with the information contained in other applicable regulations.

- Resource Conservation and Recovery Act (RCRA) (USC, 1976a) - This act authorizes the EPA and the States to regulate hazardous and solid wastes.
- Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) (USC, 1980) and 40 CFR 300 (EPA, 1992c) - This act requires the identification and cleanup of inactive hazardous waste sites by responsible parties, and imposes certain response and reporting requirements for releases of hazardous substances.
- Superfund Amendments and Reauthorization Act (SARA) (USC, 1986) and 40 CFR 300 (EPA, 1992c).

Interagency agreements can also exist between DOE, EPA, state, and local agencies (Daugherty, 1993). Any special arrangement agreed to as part of an interagency agreement will need to be honored during the D&D activities.

### **10.1.2 Residual Radioactivity Levels**

A primary concern in the D&D of any nuclear facility is the level of residual radioactivity that may be permitted for unrestricted use. However, the emphasis of this document is on occupational radiological protection. See Section 4.2.4 for guidance on contamination monitoring in the workplace. Additional information on acceptable residual levels may be found in the following sources. This list is not inclusive and facilities must determine the applicable requirements. The U.S. Nuclear Regulatory Commission (NRC) in Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (AEC, 1974), and draft ANSI N13.12 (ANSI, 1988b), provide definitive values for acceptable surface contamination levels for termination of operating licenses for nuclear reactors and for materials, equipment, and facilities. Both of these documents are based on the outdated methodology of ICRP Publication 2 (ICRP, 1959) and not the currently used

methodology of ICRP Publication 26 (ICRP, 1977) and Publication 30 (ICRP, 1979); therefore, they require updating (see Kennedy and Strenge, 1992). In addition, these documents make no mention of other limits, such as limits for soil contamination or volume sources.

The EPA has been mandated by Congress to develop guidelines that will be applicable to all nuclear facilities as well as to the release of formerly contaminated or controlled radioactive facilities for unrestricted release. Such guidelines will likely be based on the radiation dose to the maximum exposed member of the general population. The maximum allowable annual dose has not yet been determined, but values of 50, 10, 1, and 0.1 mrem/y are being considered by the EPA as the “de facto de minimis” levels for the disposal of contaminated material.

Section 4 of DOE Order 5400.5, Radiation Protection of the Public and Environment (DOE, 1990b), provides the following DOE guidelines for cleanup of residual radioactive material, management of the resulting wastes, and release of property. The basic public dose limits for exposure to residual radioactive material in addition to natural background exposures is a 100-mrem (1-mSv) effective dose equivalent in a year. The effective dose equivalent in a year is the sum of the effective dose equivalent from exposures to radiation sources external to the body during the year plus the cumulative effective dose equivalent (CEDE) from radionuclides taken into the body during the year. See DOE/CH-8901, A Manual for Implementing Residual Radioactive Material Guidelines (DOE, 1989a), for procedures for deriving specific property guidelines for allowable levels of residual radioactive material, based on the dose limit of 100 mrem (1 mSv).

DOE Order 5400.5 (DOE, 1990b) also provides the following guidelines for 1) residual concentrations of radionuclides in soil, 2) concentrations of airborne radon decay products, 3) external gamma radiation, 4) surface contamination, and 5) radionuclide concentrations in air or water:

- **Residual radionuclides in soil** - Generic guidelines for thorium and radium ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{230}\text{Th}$ , and  $^{232}\text{Th}$ ) are 5 pCi/g averaged over the first 15 cm of soil below the surface and 15 pCi/g averaged over 15-cm-thick layers of soil more than 15 cm below the surface. For other radionuclides in soil (e.g., plutonium),

specific guidelines should be derived from the basic dose limit by means of an environmental pathway analysis using specific property data where available. Residual concentrations of radioactive material in soil are defined as those in excess of background concentrations averaged over an area of 100 m<sup>2</sup>.

- **Airborne radon decay products** - Applicable generic guidelines are found in 40 CFR 192 (EPA, 1992b). In any occupied or habitable building, the objective of remedial action should be, and a reasonable effort should be made to achieve, an annual average (or equivalent) radon decay product concentration (including background) not to exceed 0.02 WL. Remedial actions by DOE are not required to comply with this guideline when there is reasonable assurance that residual radioactive material is not the source of the radon concentration.
  
- **External gamma radiation** - The average level of gamma radiation inside a building or habitable structure on a site to be released without restrictions should not exceed the background level by more than 20 µR/h.
  
- **Surface contamination** - The DOE guidelines on transuranic surface contamination levels are given in a DOE memorandum dated November 17, 1995, "Application of DOE 5400.5 requirements for release and control of property containing residual radioactive materials," the guideline values are as follows:

Guidelines

Removable Contamination	20 dpm/100 cm <sup>2</sup>
Total (Fixed plus Removable Contamination)	500 dpm/100 cm <sup>2</sup>

- **Residual radionuclides in air and water** - Residual concentrations of radionuclides in air shall not cause members of public to receive an effective dose equivalent greater than 10 mrem (0.1 mSv) in one year [DOE Order 5400.5 (DOE, 1990b)]. In 40 CFR 141, National Primary Drinking Water Regulations (EPA, 1992a), the EPA provides a limit of 4 mrem/y annual dose equivalent to the whole body or any internal organ of any member of the public from manmade radionuclides in drinking water.

The NRC is updating their decommissioning regulations and criteria. The new NRC methodology is presented in NUREG/CR-5512 (Kennedy and Streng, 1992) and is consistent with the recommendations in ICRP Publications 26 and 30 (1977, 1979). The NRC will establish a three-layered hierarchy for developing decommissioning criteria to be used to evaluate the release of property after D&D. The first and second layers use conservative models and parameters, and the third layer uses site-specific models and data to provide a more accurate approximation of actual conditions. Four criteria will be calculated upon which to make D&D decisions: 1) a surface contamination level for buildings and building materials (in dpm/100 cm<sup>2</sup>), 2) volume contamination criteria for volume sources in buildings (in pCi/g), 3) soil contamination criteria (in pCi/g), and 4) a total site inventory (in Ci).

These criteria require calculation of dose to members of the general population. The scenarios for exposure will have to include all exposure pathways that are credible under the proposed disposition. If the site is part of a closely guarded government reservation, certain pathways may be eliminated, such as the use of well water directly from the site and ingestion of significant quantities of fruits and vegetables grown on the site. However, if the site will be released for unrestricted use, such scenarios should be considered. The computer codes used for calculation of dose to the public from decommissioned facilities will include the currently accepted exposure models and site-specific or maximum credible parameters for exposure pathways.

Finally it should be noted that a multi-agency effort is currently proceeding to develop measurement and decision criteria applicable to D&D projects. The Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) has been drafted and

provided for public comment in December 1996 (NRC 1996). When MARSSIM is finalized, it should provide detailed techniques applicable to the D&D of DOE facilities.

## **10.2 DESIGN FEATURES**

Design of the facility should allow easy D&D of equipment and materials. Details on designing facilities for ease of decommissioning are found in DOE Order 6430.1A, General Design Criteria (DOE, 1989b) and are discussed in the following sections. 10 CFR 835.1002 and Appendix C of this document provide additional guidance on facility design.

### **10.2.1 Building Materials**

In general, the design features that aid in contamination control during operation also facilitate decommissioning. The inclusion of all the building materials suggested in this section may be cost-prohibitive, but they should be considered if the budget allows. The maintenance procedures that are used during operation are also important in controlling the spread of contamination to clean areas and, therefore, they facilitate decommissioning, too.

Less permeable building materials are more easily decontaminated. Any concrete with uncoated surfaces that comes in contact with plutonium solutions or plutonium-contaminated air will require surface removal and disposal as radioactive waste at the end of its life. If there are cracks through which contaminated solutions have penetrated, the entire structure may need to be disposed of as radioactive waste.

Metal surfaces may also require decontamination. In general, the more highly polished the surface, the easier it will be to decontaminate. If feasible, all stainless steel that will come into contact with plutonium should be electropolished before being placed into service. If high-efficiency particulate air (HEPA) filtration has failed at any time during facility operation, roofs may require decontamination. Metal roofs are easiest to decontaminate, but even these may contribute to the volume of radioactive waste unless unusual measures are taken to clean them. Built-up and composition roofs will be difficult to clean to unrestricted release levels.

Interior surfaces are most easily cleaned if they were completely primed and painted before the introduction of radioactive materials into the facility. If interior surfaces are repainted during operation, their disposal as clean waste is likely to require removal of the paint. However, if the paint has deteriorated, cleaning for unrestricted use may be as difficult as if the material had never been painted. Wood will almost certainly become contaminated, as will plasterboard and other such materials.

Floor surfaces are likely to be a problem. Concrete should be well sealed and covered with a protective surface. Single sheet, vinyl flooring with heat-sealed seams is preferable to asphalt or vinyl tile because it is more easily cleaned. If the floor needs resurfacing, it is preferable to overlay new flooring material rather than remove the old material and expose the underlying floor.

Carpets are not recommended because they are difficult to clean and survey and bulky to dispose of and they do not adequately protect the underlying surface. In some areas, such as control rooms, their use may be justified by noise control requirements; however, their contamination control limitations should be considered. If used, carpets should be surveyed frequently and disposed of as radioactive waste when they become contaminated.

### **10.2.2 Ventilation Systems**

In addition to decommissioning considerations, the design of the ventilation system will depend on the operations that will be conducted in the facility. Adequate air flow for all operations and good design practices will help keep the facility clean during operations and will facilitate decommissioning. Fiberglass duct work may present a fire hazard and may be more difficult to decontaminate than stainless steel, especially stainless steel that has been electropolished. Welded joints are less likely to collect contamination than bolted ones; however, bolted joints are easier to remove and the most contaminated areas are readily accessible for cleaning.

Filters should be positioned in ventilation systems to minimize contamination of ductwork (e.g., filtration of glove-box exhaust air before it enters a duct leading to a plenum).

### **10.2.3 Piping Systems**

Potentially contaminated piping systems that are imbedded in concrete are a common and relatively expensive decommissioning problem. Most often, they must be sealed and removed last, after all other radioactive material has been removed and the building is being demolished by conventional methods. Often, they provide the major impetus for demolishing a building rather than converting it to some non-nuclear use. For this reason, it is best to run pipes in chases or tunnels that have been lined (usually with stainless steel) to prevent contamination from penetrating building surfaces. To minimize hand jackhammer work required during decommissioning, floor drains should not be enclosed in concrete.

### **10.2.4 Soil-Contamination Considerations**

Depending on the activity levels found, locations where contaminated effluents have penetrated the ground may require excavation during decommissioning. The facility design should minimize such areas. Particular attention should be paid to storm runoff from roofs, storage areas, contaminated equipment storage, and liquid waste treatment impoundments (including sanitary sewage systems if they may receive some small amount of contamination during the life of the facility.)

### **10.2.5 Other Features**

Installed decontamination and materials-handling equipment that facilitates operation and maintenance generally facilitates decommissioning in two ways. First, it can be used for its intended purposes of cleaning and moving equipment during the decommissioning phase. Even more important, it usually contributes to a cleaner, better maintained facility, where nonfunctional equipment is moved out when it is no longer needed and work surfaces are kept free of spreadable contamination.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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Other features discussed in DOE Order 6430.1A, General Design Criteria (DOE, 1989b) include the following:

- Minimizing service piping, conduits, and ductwork;
- caulking or sealing all cracks, crevices, and joints;
- using modular, separable confinements for radioactive or other hazardous materials to preclude contamination of fixed portions of the structure;
- using localized liquid transfer systems that avoid long runs of buried contaminated piping;
- using equipment that precludes the accumulation of radioactive or other hazardous materials in relatively inaccessible areas, including curves and turns in piping and ductwork;
- using designs that ease cut-up, dismantling, removal, and packaging of contaminated equipment from the facility;
- using modular radiation shielding, in lieu of or in addition to monolithic shielding walls;
- using lifting lugs on large tanks and equipment; and
- using fully drainable piping systems that carry contaminated or potentially contaminated liquids.

### **10.3 DECONTAMINATION AND DECOMMISSIONING PROGRAM REQUIREMENTS**

The basic requirements for a D&D program at a DOE facility are found in Section 5 of DOE Order 5820.2A, Radioactive Waste Management (DOE, 1988b). Planning for facility decommissioning should be initiated during the design phase for new facilities and before



termination of operations for existing operational facilities (see Section 5,3.a.(2), of DOE Order 5820.2A). DOE Order 5820.2A divides the discussion of requirements into several time periods: pre-operational and operational activities, post-operational activities, D&D activities, and post-decommissioning activities. To assist in D&D activity planning the Office of Environmental Management distributed the "Decommissioning Resource Manual." Refer to that document for guidance.

Requirements relating to occupational radiological protection include (this is not an all inclusive list, facilities must determine the applicable set of requirement):

DOE Order 4700.1, Project Management System 4.a (DOE, 1992e), contains the requirements by which all DOE projects must be managed; Section 2, A.3.c, requires that a project management plan be developed for major system acquisitions and major projects; Item 2.d of Attachment II-4 states that environment, safety, and health technical requirements for project design and implementation should be included in the work-plan section of the project management plan.

In addition, Section 5, 3.d.(3) of DOE Order 5820.2A (DOE, 1988b) states that a decommissioning project plan shall be prepared according to DOE Order 4700.1 and shall include the following:

- development of a health and safety plan for decommissioning and
- projections of occupational exposure;

Safety analysis reports (SARs) typically do not need to be written to cover D&D activities for those facilities that are currently shut down or will be shut down in the near future [see DOE Order 5480.23, Nuclear Safety Analysis Reports (DOE, 1992d)]. Only a decommissioning plan would be required. (However, a "Bases" for Interim Operations is required for facilities not yet shut down to continue operations.) The requirement for a decommissioning plan will apply for a large number of the plutonium-contaminated facilities that are awaiting decommissioning. For new or operating facilities, provisions for D&D need to be included in the new or updated SAR.

## **10.4 DECONTAMINATION AND DECOMMISSIONING TECHNIQUES**

This section concentrates on decontamination techniques to be used in the final decommissioning of a plutonium-contaminated facility for unrestricted release. Some of these techniques are similar to those used during routine operations (e.g., some equipment and building surface decontamination). Contamination detection methods are similar for routine and D&D operations and are discussed in Chapter 4.

### **10.4.1 Equipment and Surface Decontamination**

Decontamination of surface areas may be as simple as hosing off the floors with water, washing surfaces with detergent and water, or wiping with household dust cloths. Waste material generated from decontamination activities (e.g., water and wipe material) must be contained and disposed of as radioactive waste. For some locations, vacuuming the surfaces may be appropriate. If vacuuming is used, HEPA-filtered vacuum systems are required to keep airborne radioactive material out of the vacuum exhaust.

For some operations, periodic surface flushing with water may be adequate to maintain acceptable contamination levels. Precautions should ensure control and collection of run-off water so that material may be recovered and waste water analyzed before discharge. Depending upon which isotope of plutonium is involved, geometrically safe containers may be required for collecting and holding the liquid.

Depending upon the physical and chemical form of the plutonium and the type of surface, plutonium may become imbedded in the surface. Removal of embedded material may require physical abrasion, such as scabbling, grinding, sand blasting, or chipping, or it may be accomplished using chemical etching techniques. If the surface is porous, complete replacement could be necessary. The use of high-pressure water (hydroblasting) has been quite successful for metal and concrete surfaces.

Ultrasonic cleaning techniques (electropolishing) or chemical baths may be useful for decontamination of high-cost items if the chemicals used are compatible with the material to be cleaned.

A description of different decontamination techniques is found in DOE/EV/10128-1, DOE Decommissioning Handbook (DOE, 1980), and publications by Allen (1985) and the Electric Power Research Institute (EPRI, 1989). The DOE Decommissioning Handbook also includes guidance on decontamination techniques, assessment of environmental impacts, disposition of wastes, and preparation of decommissioning cost estimates.

## **10.5 DECONTAMINATION AND DECOMMISSIONING EXPERIENCE**

Considerable experience has been gained in D&D of commercial plutonium facilities, as discussed in Hoovler et al. (1986), Denero et al. (1984), and Adams et al. (1982). Hoovler et al. (1986) discuss the decommissioning programs carried out at two Babcock and Wilcox buildings in Lynchburg, Virginia, which housed plutonium/uranium fuel development laboratories. They include information on decommissioning and quality assurance plans, conducting D&D work, performing radiological surveys before and after D&D work, and disposing of the waste. Denero et al. (1984) discuss the D&D of the Westinghouse Nuclear Fuel Facility at Cheswick, Pennsylvania. They describe the facility and its operations, nondestructive assay techniques, equipment required for dismantling and packaging the waste, and management of the TRU waste. Adams et al. (1982) discuss the complete D&D of the Westinghouse Advanced Reactors Division Fuel Laboratories at the Cheswick, Pennsylvania, site. The report describes the D&D plans, the EA written for the operation, the quality assurance plan, and the health physics, fire control, and site emergency manuals written for the operation.

Discussions of D&D activities at several DOE plutonium facilities are provided by Adkisson (1987), Bond et al. (1987), and King (1980), as well as by Shoemaker and Graves (1980), Garner and Davis (1975), Wynveen et al. (1982), Hunt et al. (1990), Freas and Madia (1982), and Garde et al. (1982a, 1982b). They describe D&D activities that took place in several types of plutonium facilities, including fabrication facilities, research and development laboratories, and a storage facility. Plutonium-contaminated glove boxes, hoods, ventilation ductwork, laboratory equipment, structural components (i.e., walls and floors), and filter banks were decontaminated. Typically, decontamination methods included wiping with a damp cloth or mop, using strippable coatings, mechanical spalling of concrete floor surfaces, and fixating contamination on a piece of equipment (e.g., a hood), followed by disassembling the item inside a contamination control enclosure.

Some lessons learned from past studies include the following:

- Waste management planning should begin early in the D&D planning stages and consider the following:
  - The possibility exists that there may be more stringent regulations for shipping hazardous or radioactive wastes than disposing of it and
  - Compliance with all applicable waste management requirements may be difficult (e.g., the Waste Isolation Pilot Project (WIPP) does not accept mixed wastes or TRU waste that contains plastics or foams).
- It is difficult to decontaminate some items with inaccessible surfaces to less than the TRU limit (100 nCi/g) so that they can be disposed of as LLW. In some situations, it may be possible to decontaminate to <100 nCi/g of TRU, but the decontamination process may generate a large volume of liquid waste or be time-consuming enough to prohibit its use.
- Temporary enclosures are effective in controlling contamination when reducing the size of large equipment such as glove boxes. Any loose contamination on the equipment should be fixed before placing it in the enclosure.
- Criticality safety issues regarding the geometry of any waste material containing fissile material need to be considered.

Adkisson (1987) reported on the decommissioning of a plutonium fuel fabrication plant at the Sequoyah Fuels Corporation's Cimarron Facility, located in north-central Oklahoma. Process equipment, glove boxes, tanks, piping, and ventilation ducts required decontamination. Controlling personnel exposures, maintaining containment of radioactive material during the dismantling of contaminated items, and reducing the volume of TRU material were the primary considerations during the decommissioning activities. A large modified glove box provided containment for dismantling and cutting up the various equipment using a plasma-arc unit. A passive, gamma-ray nondestructive assay technique (heavily shielded NaI detector with collimator) was used to measure the plutonium content of cut-up pieces. Finally, the loaded

waste drums were measured using a waste drum counter to ensure that plutonium levels were less than 100 nCi/g.

A number of plutonium-contaminated facilities have been decommissioned at Mound Laboratory (Bond et al., 1987). Interdepartment management teams, including representatives from program management, operations, project engineering, maintenance, technical support, and environmental, safety, and health were established for the D&D projects. The team met monthly to discuss program status and they met quarterly with DOE staff. A graded D&D approach was used. First, standard cleaning (e.g., wiping with a damp cloth) and flushing techniques were used to remove loose contamination. Then, more aggressive decontamination methods were performed inside temporary enclosures. Finally, glove boxes and equipment that could not be decontaminated to unrestricted release levels were cut into sections using a plasma-cutting method and then packaged as waste. The plasma-cutting method generated less smoke, thus reducing the particulate accumulation on the HEPA filters.

During cleanup of a plutonium-contaminated storage facility, strippable fixatives were used as a contamination control and a decontamination method (King, 1980). Fixatives in combination with cheese cloth were used to clean smooth vertical surfaces and difficult-to-reach areas. The cheese cloth was placed on the area to be cleaned and then sprayed with a fixative. The cheese cloth and fixative were then stripped from the surface, removing contamination in the process. Accidental criticalities can be a concern when disposing of this material that contains fissile material contamination, as discussed in Section 8.0, and criticality safety specialists should be consulted. Facility personnel also need to determine if the fixative is classified as a hazardous material and dispose of it accordingly.

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## APPENDIX A

### GLOSSARY

**abnormal situation:** Unplanned event or condition that adversely affects, potentially affects or indicates degradation in the safety, security, environmental or health protection performance or operation of a facility. **(RCM)**

**absorbed dose:** The energy absorbed by matter from ionizing radiation per unit mass of irradiated material at the place of interest in that material. The absorbed dose is expressed in units of rad (or gray) (1 rad = 0.01 gray). **(10 CFR 835)**

**activity median aerodynamic diameter:** The diameter of a sphere having a density of  $1 \text{ g cm}^{-3}$  with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol. **(Internal Dosimetry IG)**

**air sampling:** A form of air monitoring in which an air sample is collected and analyzed at a later time, sometimes referred to as retrospective air monitoring.

**air monitoring:** Actions to detect and quantify airborne radiological conditions by the collection of an air sample and the subsequent analysis either in real-time or off line laboratory analysis of the amount and type of radioactive material present in the workplace atmosphere. **(Internal Dosimetry IG)**

**airborne radioactive material:** Radioactive material in any chemical or physical form that is dissolved, mixed, suspended, or otherwise entrained in air.

**airborne radioactivity area:** Any area where the measured concentration of airborne radioactivity, above natural background, exceeds or is likely to exceed 10% of the derived air concentration (DAC) values listed in appendix A or appendix C of 10 CFR 835. **(10 CFR 835)**

**alarm set point:** The count rate at which a continuous air monitor will alarm, usually set to correspond to a specific airborne radioactive material concentration by calculating the sample medium buildup rate.

**ambient air:** The general air in the area of interest (e.g., the general room atmosphere) as distinct from a specific stream or volume of air that may have different properties.

**annual limit on intake (ALI):** The derived limit for the amount of radioactive material taken into the body of an adult worker by inhalation or ingestion in a year. ALI is the smaller value of intake of a given radionuclide in a year by the reference man (ICRP Publication 23) that would result in a committed effective dose equivalent of 5 rem (0.05 sievert) or a committed dose equivalent of 50 rem (0.5 sievert) to any individual organ or tissue. ALI values for intake by ingestion and inhalation of selected radionuclides are based on Table 1 of the U.S. Environmental Protection Agency's Federal Guidance Report No. 11, Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion, published September 1988. **(10 CFR 835)**

**as low as is reasonably achievable (ALARA):** The approach to radiation protection to manage and control exposures (both individual and collective) to the work force and to the general public to as low as is reasonable, taking into account social, technical, economic, practical, and public policy considerations. ALARA is not a dose limit but a process which has the objective of attaining doses as far below the applicable limits of 10 CFR 835 as is reasonably achievable. **(10 CFR 835)**

**bioassay:** The determination of kinds, quantities, or concentrations, and, in some cases, locations of radioactive material in the human body, whether by direct measurement or by analysis, and evaluation of radioactive materials excreted or removed from the human body. **(10 CFR 835)**

**breathing zone air monitoring:** Actions conducted to detect and quantify the radiological conditions of air from the general volume of air breathed by the worker, usually at a height of 1 to 2 meters. See *personal air monitoring*. **(Workplace Air Monitoring IG)**

**contamination area:** Any area where contamination levels are greater than the values specified in appendix D of 10 CFR 835, but less than or equal to 100 times those levels. **(10 CFR 835)**

**continuous air monitor (CAM):** An instrument that continuously samples and measures the levels of airborne radioactive materials on a "real-time" basis and has alarm capabilities at preset levels. **(10 CFR 835)**

**contractor:** Any entity under contract with the Department of Energy with the responsibility to perform activities at a DOE site or facility. **(10 CFR 835)**

**controlled area:** Any area to which access is managed in order to protect individuals from exposure to radiation and/or radioactive material. Individuals who enter only the controlled area without entering radiological areas are not expected to receive a total effective dose equivalent of more than 100 mrem (0.001 sievert) in a year. **(10 CFR 835)**

**decontamination:** The process of removing radioactive contamination and materials from personnel, equipment or areas. **(RCM)**

**Department of Energy operations:** Those activities funded by DOE for which DOE has authority to enforce for environmental protection, safety, and health protection requirements. **(DOE Order 5484.1)**

**Department of Energy site:** Either a tract owned by DOE or a tract leased or otherwise made available to the Federal Government under terms that afford to the Department of Energy rights of access and control substantially equal to those that the Department of Energy would possess if it were the holder of the fee (or pertinent interest therein) as agent of and on behalf of the Government. One or more DOE operations/program activities are carried out within the boundaries of the described tract. **(DOE Order 5484.1)**

**derived air concentration (DAC):** For the radionuclides listed in appendix A of 10 CFR 835, the airborne concentration that equals the ALI divided by the volume of air breathed by an average worker for a working year of 2000 hours (assuming a breathing volume of 2400 m<sup>3</sup>). For the radionuclides listed in appendix C of this part, the air immersion DACs were calculated for a continuous, non-shielded exposure via immersion in a semi-infinite atmospheric cloud. The value is based upon the derived airborne concentration found in Table 1 of the U.S. Environmental Protection Agency's Federal Guidance Report No. 11, Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion, published September 1988. **(10 CFR 835)**

**detector:** A device or component that produces a measurable response to ionizing radiation. **(Portable Instrument Calibration IG)**

**DOELAP:** The Department of Energy Laboratory Accreditation Program for personnel dosimetry .  
(RCM)

**dose:** The amount of energy deposited in body tissue due to radiation exposure. (RCM)

**exposure:** The general condition of being subjected to ionizing radiation, such as by exposure to ionizing radiation from external sources or to ionizing radiation sources inside the body. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air.  
(Internal Dosimetry IG)

**fissionable materials:** A nuclide capable of sustaining a neutron - induced fission chain reaction (e.g., uranium-233, uranium-235, plutonium-238, plutonium 239, plutonium -241, neptunium-237, americium-241 and curium-244) (10 CFR 830).

**fixed contamination:** Any area with detectable removable contamination less than the removable contamination values of Appendix D of 10 CFR 835 and fixed contamination at levels that exceed the total contamination values of Appendix D of 10 CFR 835. (Posting and Labeling IG)

**fixed-location sampler:** An air sampler located at a fixed location in the workplace.

**grab sampling:** A single sample removed from the workplace air over a short time interval, typically less than one hour.

**hazardous waste:** Because of its quantity, concentration, and physical, chemical, or infectious characteristics, hazardous waste may cause or significantly contribute to an increase in mortality, or an increase in serious irreversible or incapacitating reversible illness; it may pose a potential hazard to human health or the environment when improperly treated, stored, transported, disposed of, or otherwise managed. (DOE/S-0101)

**high contamination area:** Any area where contamination levels are greater than 100 times the values specified in appendix D of 10 CFR 835. (10 CFR 835)

**high-efficiency particulate air (HEPA) filter:** Throwaway extended pleated medium dry-type filter with 1) a rigid casing enclosing the full depth of the pleats, 2) a minimum particle removal efficiency of 99.97% for thermally generated monodisperse di-octyl phthalate smoke particles with a diameter of 0.3  $\mu\text{m}$ , and 3) a maximum pressure drop of 1.0 in. w.g. when clean and operated at its rated airflow capacity. **(RCM)**

**high radiation area:** Any area, accessible to individuals, in which radiation levels could result in an individual receiving a deep dose equivalent in excess of 0.1 rem (0.001 sievert) in 1 hour at 30 cm from the radiation source or from any surface that the radiation penetrates. **(10 CFR 835)**

**HLW:** High-level waste (HLW) is the material that remains following the reprocessing of spent nuclear fuel and irradiated targets from reactors. The HLW is highly radioactive and generates heat on its own. Some of its elements will remain radioactive for thousands of years. Because of this, HLW must be managed very carefully and all handling must be performed from behind heavy protective shielding. **(DOE/S-0101)**

**intake:** The amount of radionuclide taken into the body by inhalation, absorption through intact skin, injection, ingestion or through wounds. Depending on the radionuclide involved, intakes may be reported in mass (e.g.,  $\mu\text{g}$ , mg) or activity (e.g.,  $\mu\text{Ci}$ , Bq) units. **(Internal Dosimetry IG)**

**LLW:** Low-level waste (LLW) is any radioactive waste that is not HLW, spent nuclear fuel, TRU waste, or uranium mill tailings. The LLW is typically contaminated with small amounts of radioactivity dispensed in large amounts of material. The LLW is generated in every process involving radioactive materials in the DOE including decontamination and decommissioning projects. **(DOE/S-0101)**

**minimum detectable amount/activity (MDA):** The smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability B of non-detection (Type II error) while accepting a probability  $\alpha$  of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error). **(ANSI N13.30-1996)**

**MW:** Mixed waste (MW) is waste that contains both radioactive and hazardous wastes. Any of the types of radioactive waste described can be a mixed waste if it contains any hazardous wastes. In fact, all of

DOE's HLW is mixed waste because of the chemicals used to reprocess the fuel that resulted in the generation of the material or because it is suspected to contain hazardous materials. **(DOE/S-0101)**

**occupational exposure:** An individual's exposure to ionizing radiation (external and internal) as a result of that individual's work assignment. Occupational exposure does not include planned special exposures, exposure received as a medical patient, background radiation, or voluntary participation in medical research programs. **(10 CFR 835)**

**personal air monitoring:** The monitoring of air for radioactive particles in the immediate vicinity of an individual radiation worker's nose and mouth, usually by a portable sampling pump and collection tube (such as a lapel sampler) worn on the body. Personal air monitoring is a special case of breathing zone air monitoring. **(Workplace Air Monitoring IG)**

**portable air sampler:** An air sampler designed to be moved from area to area.

**radiation area:** Any area accessible to individuals in which radiation levels could result in an individual receiving a deep dose equivalent in excess of 0.005 rem (0.05 millisievert) in 1 hour at 30 cm from the source or from any surface that the radiation penetrates. **(10 CFR 835)**

**radiation-generating device (RDG):** The collective term for devices which produce ionizing radiation, sealed sources which emit ionizing radiation, small particle accelerators used for single purpose applications which produce ionizing radiation (e.g., radiography), and electron-generating devices that produce x-rays incidentally. **(Radiation-Generating Devices IG)**

**radioactive material:** For the purposes of the Radiological Control Manual, radioactive material includes any material, equipment or system component determined to be contaminated or suspected of being contaminated. Radioactive material also includes activated material, sealed and unsealed sources, and material that emits radiation. **(RCM)**

**radioactive material area:** An area where radioactive material is used, handled, or stored. **(Posting and Labeling IG)**

**radiological area:** Any area within a controlled area which must be posted as a "radiation area," "high radiation area," "very high radiation area," "contamination area," "high contamination area," or "airborne radioactivity area" in accordance with 10 CFR 835.6093. **(10 CFR 835)**

**radiological worker:** A general employee whose job assignment involves operation of radiation-producing devices or working with radioactive materials, or who is likely to be routinely occupationally exposed above 0.1 rem (0.001 sievert) per year total effective dose equivalent. **(10 CFR 835)**

**radiological work permit (RWP):** The permit that identifies radiological conditions, establishes worker protection and monitoring requirements, and contains specific approvals for radiological work activities. The Radiological Work Permit serves as an administrative process for planning and controlling radiological work and informing the worker of the radiological conditions. **(RCM)**

**radiological protection organization:** A contractor organization responsible for radiation protection activities within contractor facilities. This organization is independent of the line organizational element responsible for production, operation, or research activities and should report to the contractor senior site executive. **(Sealed Source IG)**

**real-time air monitoring:** Collection and real-time analysis of the workplace atmosphere using continuous air monitors (CAMs).

**refresher training:** The training scheduled on the alternate year when full retraining is not completed for Radiological Worker I and Radiological Worker II personnel. **(RCM)**

**removable contamination:** Radioactive material that can be removed from surfaces by nondestructive means, such as casual contact, wiping, brushing or washing. **(RCM)**

**representative air sampling:** The sampling of airborne radioactive material in a manner such that the sample collected closely approximates both the amount of activity and the physical and chemical properties (e.g., particle size and solubility) of the aerosol to which the workers may be exposed.

**residual radioactive material:** Any radioactive material which is in or on soil, air, water, equipment, or structures as a consequence of past operations or activities. **(Draft 10 CFR 834)**



**sanitary waste:** Sanitary waste is waste that is neither hazardous nor radioactive. (DOE/S-0101)

**source-specific air sampling:** Collection of an air sample near an actual or likely release point in a work area using fixed-location samplers or portable air samplers.

**survey:** An evaluation of the radiological conditions and potential hazards incident to the production, use, transfer, release, disposal, or presence of radioactive material or other sources of radiation. When appropriate, such an evaluation includes a physical survey of the location of radioactive material and measurements or calculations of levels of radiation, or concentrations or quantities of radioactive material present. (10 CFR 835)

**TRU:** Transuranic (TRU) waste refers to waste materials containing elements with atomic numbers greater than 92. These elements are generally alpha-emitting radionuclides that decay slowly. The TRU waste contains a concentration of these elements greater than 100 nCi/g. The TRU waste is not as intensely radioactive as HLW. The TRU waste also decays slowly, requiring long-term isolation. (DOE/S-0101)

**very high radiation area:** Any area accessible to individuals in which radiation levels could result in an individual receiving an absorbed dose in excess of 500 rad (5 gray) in 1 hour at 1 meter from a radiation source or from any surface that the radiation penetrates. (10 CFR 835)

**workplace monitoring:** The measurement of radioactive material and/or direct radiation levels in areas that could be routinely occupied by workers.

## APPENDIX B

### PLUTONIUM IN DEPARTMENT OF ENERGY FACILITIES

The mission of the DOE complex has changed significantly from the operation of the many and varied facilities required for the production of a nuclear deterrent system to maintenance of a smaller-scale nuclear weapons program, dismantling of surplus weapons, and decontamination and decommissioning of facilities no longer needed. Many, although not all, of the DOE facilities were involved in the production, separation, processing, handling, or storing of plutonium. This appendix describes those DOE facilities which were significantly involved in these activities. It provides future users of this manual with a brief description of the facilities and processes involving plutonium within the DOE complex. Personnel at facilities with similar needs are encouraged to contact one another and share information for increased safety and efficiency.

Many of the plutonium facilities given in this appendix are no longer in production and either are currently or are scheduled to be incorporated into the environmental restoration program. The following descriptions will alert the user to potential plutonium concerns within the facility.

#### B.1 ARGONNE NATIONAL LABORATORY - WEST

Research and development activities involving plutonium at the Argonne National Laboratory have been limited to Argonne West at the Idaho National Engineering Laboratory (INEL). Pu-U-Mo alloy fuel plates used in the Zero Power Physics Reactor are in storage at the facility. Some fuel-cycle research and development using plutonium has been conducted, and plutonium metal and oxides are in storage.

#### B.2 BATTELLE COLUMBUS LABORATORY - WEST JEFFERSON FACILITY

The West Jefferson Facility Nuclear Sciences area contained a hot laboratory, an administrative building, a retired research reactor, and a decommissioned plutonium laboratory. The hot laboratory was used for the examination of irradiated reactor fuel and other materials, as well as for other purposes such as the preparation of sealed radiation sources. Entire power reactor fuel assemblies were handled in this facility. The administrative building was used for critical experi-

ments from 1957 through 1963. The operating license was terminated in 1970 although a storage vault was still being used for special nuclear material storage in 1987. The research reactor ceased operation in 1975 and was being decommissioned in 1987. Since the plutonium laboratory was dismantled in 1985, only hazardous materials operations have been conducted. The facility has been undergoing decontamination and decommissioning, which should be completed in the near future.

### **B.3 FEED MATERIAL PRODUCTION CENTER**

The Feed Material Production Center was primarily a uranium processing and production facility. However, trace quantities of plutonium were found in some of the recycled feed uranium. The site is undergoing decontamination and decommissioning.

### **B.4 HANFORD SITE**

Nine reactors were operated at various times at the Hanford Site for the production of plutonium. Plutonium separation from irradiated fuel started at the Hanford Site in December 1944 in T-Plant in the 200-West Area. Processed material from T-Plant was transferred to 224-T and to 231-Z for additional processing. The final Hanford product was a plutonium nitrate paste which was shipped to Los Alamos for further processing. A sister facility, B-Plant, was constructed to increase capacity. A new facility, Redox, was started in 1952 using a new, more efficient process. Another plutonium separations facility soon followed, the PUREX Plant, in 1955. Because of the capacity of this new process and its efficiency, T-Plant, B-Plant, and Redox were shut down. Plutonium purification and further processing was conducted in 234-5 facility. A detailed history of these facilities and processes can be found in WHC-MR-0437, A Brief History of the Purex and UO<sub>3</sub> Facilities (Gerber, 1993), and in WHC-MR-0452, Dramatic Change at T Plant (Gerber, 1994). It may be noted that PUREX is also currently shut down.

A large number of support facilities were required for the plutonium separations activities. Analytical and testing laboratories and research and development facilities used in the support of the plutonium activities are located in the 200-East, 200-West, and 300 Areas. Specific buildings handling plutonium in the 300 Area include 3706 in the early years and 308, 324, 325, 329, 331 and 3720 in the later years to the current time. Building 331 used plutonium aerosols for

inhalation uptake studies. Building 326 also performed analyses and measurements on small quantities of plutonium, generally environmental levels.

There are no plutonium production or processing activities currently active at the Hanford Site.

## **B.5 LAWRENCE LIVERMORE NATIONAL LABORATORY**

The Lawrence Livermore National Laboratory conducts significant design, development, and quality control activities using plutonium. The Plutonium Facility (Building 332) was constructed in 1960 to provide a capability for safe handling and storage of plutonium in support of the laboratory's metallurgical research and development effort and the nuclear weapons design program. The facility has been expanded and upgraded several times to increase capacity and enhance safety features. All the capabilities necessary for fabrication of plutonium weapons parts and the metallurgical evaluations accompanying those operations are available within the facility. The Laser Isotope Separation program has been a major user of the facility.

## **B.6 IDAHO NATIONAL ENGINEERING LABORATORY**

The INEL use of plutonium is associated primarily with two areas. The Radioactive Waste Management Complex (RWMC) has in the past received plutonium wastes from DOE facilities such as Rocky Flats and Mound Laboratory. Plutonium waste at the RWMC is stored pending characterization and eventual disposal at the Waste Isolation Pilot Plant. Other INEL facilities using plutonium are the Argonne National Laboratory - West Site; the EBR II reactor, a fast breeder reactor fueled with plutonium and the Fuel Conversion Facility, built to demonstrate the concept of removing EBR II fuel, processing it in an enclosed attached facility, and reusing the plutonium and actinides in refueling the reactor. Closure of these facilities curtailed these functions, but the remaining fuel is still stored pending eventual disposal. Additional plutonium was used at the Zero Power Reactor for design testing. Plutonium is an incidental radioisotope encountered at the Idaho Chemical Processing Plant.

## **B.7 LOS ALAMOS NATIONAL LABORATORY**

The Los Alamos National Laboratory conducts

- nuclear weapons research, development and testing;
- plutonium chemistry and metallurgy research;
- fabrication of radioisotope thermoelectric generators;
- explosives testing involving depleted uranium;
- accelerator-based nuclear physics research and applied technologies;
- tritium research and applications;
- materials science and testing involving radioactive materials and accelerators;
- nuclear criticality experimentation;
- mixed fission product analysis including hot cell work;
- decontamination and decommissioning of large scale radiological facilities; and
- radioactive and mixed waste treatment and storage.

It also uses X-ray devices, neutron generators, and sealed sources. Generally, the site has a limited quantity of plutonium, which is handled and stored as a result of laboratory operations. However, Building TA-55 has significant quantities of stored plutonium including metal, oxides, and other residues.

## **B.8 MOUND LABORATORY**

The Mound plant manufactures neutron sources, chemical explosive detonators, explosive timers, explosive actuated transducers, explosive switches, heat sources fueled with  $^{210}\text{Po}$  and  $^{238}\text{Pu}$ , calorimeters, and some tritium-containing reservoirs. Other activities included stockpile evaluation of small explosive and electrical components and tritium-containing reservoirs, assembly and testing of radioisotopically fueled thermoelectric generators, and the separation, purification and commercial sales of a variety of stable isotopes and tritium. Prior to 1976, some work was done with unencapsulated  $^{238}\text{Pu}$ . However, after that time, all heat source fabrication was done with encapsulated  $^{238}\text{Pu}$ . Since its beginning in 1954, the isotopic heat source programs have produced numerous small and large General Purpose Heat Sources for space applications, including the Apollo lunar missions, and cardiac pacemakers. Limited activity is still in progress.

## **B.9 NEVADA TEST SITE**

The Nevada Test Site has been used extensively for testing of nuclear weapons, both surface and underground, as well as other related types of testing. Except for weapons components and the residue of the testing programs, no plutonium has been handled, processed, or stored at the site. A device assembly facility has been constructed on the site but has not yet been used. It would handle plutonium only as prefabricated components.

## **B.10 PANTEX PLANT**

The Pantex Plant is responsible for fabricating chemical high explosive components for nuclear weapons; disassembling nuclear weapons that have been retired from the stockpile; assembling nuclear weapons for the nation's stockpile; and maintaining and evaluating nuclear weapons in the stockpile. Currently, the major effort at the site is disassembly of weapons, with approximately 1500 being disassembled per year. Some weapons assembly is continuing, but on a relatively small scale. Large quantities of plutonium pits are normally in interim storage at the site.

## **B.11 ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE**

The mission of the Rocky Flats Environmental Technology Site (originally the Rocky Flats Plant) has changed from production of nuclear weapons components to radioactive material storage, environmental restoration, and waste management.

Plutonium activities at the site were limited to chemical and mechanical processing of plutonium and did not include plutonium production. Processing, recovering, machining, and fabricating involved primarily four buildings: 991, 707, 771, and 776/777. Two buildings, 559 and 779, were used for laboratory analysis and research. Building 371 was built as a replacement for 771 but has not been reliably utilized in that capacity. It does have analytical laboratory capability and a large storage area for plutonium.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- Building 991 is the oldest building involved in plutonium activities. However, it has recently been used only for storage and shipping/receiving of complete plutonium components (pits) to and from Pantex.
- Building 707 was used for the fabrication and assembly of nuclear weapons components. This involved melting, blending, casting, machining, fabricating, and assembling plutonium components. Building 776 had been used for these operations until 1969.
- Building 771 was used for chemical recovery and processing of plutonium to produce usable product material.
- Building 776/777 was used for disassembly of returned components from the field. This building also performed electrorefining for the separation of americium from plutonium.
- Building 559 was used as an analytical laboratory to support weapons component production, i.e., quality control, quality assurance, and other laboratory activities.
- Building 779, also a laboratory facility, provided chemical and metallurgical research and development.

The plutonium facilities at Rocky Flats have had several major contamination incidents involving plutonium fires. Since plutonium pits were the primary plutonium product shipped from the site, all other plutonium metal was considered in process and not packaged for long-term storage. Relatively large quantities of plutonium metal and oxides are located in process lines and in storage locations.

## **B.12 SANDIA NATIONAL LABORATORIES**

The Sandia National Laboratories engages in the following activities:

- nuclear weapons research, development, and testing;
- explosives testing involving radioactive materials;
- accelerator-based nuclear physics research, materials testing, and applied technologies;

- reactor-based nuclear physics research and materials testing;
- hot cell work to support analysis of fuel and material components;
- use of X-ray devices, neutron generators, and sealed sources;
- decontamination and decommissioning of radiological facilities;
- environmental restoration;
- radioactive and mixed waste treatment and storage; and
- proposed thermal battery and medical radioisotope production.

The Tonapah Test Range was established in 1957 for testing nuclear weapons ballistics, parachutes, and other non-nuclear functions for Sandia National Laboratories. Fourteen potential release areas in need of characterization have been identified.

This site has been used for various tests for which a remote site was needed for safety or security reasons. These tests have involved beryllium, uranium, and plutonium. Plutonium is the greatest concern, with three areas fenced due to plutonium contamination from dispersal tests.

#### **B.13 SAVANNAH RIVER SITE**

The Savannah River Site has extensive operations involving plutonium. Both F and B Areas contain facilities that process irradiated fuel and recover plutonium. Additional facilities provide analytical and measurement capabilities. Both  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  have been handled and stored at the site.

#### **B.14 VALLICITOS NUCLEAR CENTER**

The Vallicitos Nuclear Center is a commercially owned research site near San Francisco which is no longer active. From 1962 through 1979, the site was used for mixed oxide fuel fabrication for the DOE. A hot cell onsite was used to examine uranium fuel and reactor components and was later converted to study mixed oxide (uranium and plutonium) fuel cell rods. Decontamination is scheduled to begin in 1997.

#### **B.15 WEST VALLEY DEMONSTRATION PROJECT OFFICE**



The West Valley Demonstration Project is on the site of the only commercial nuclear fuel reprocessing plant to operate in the United States. Most of the material processed was irradiated metallic uranium fuel from the Hanford N-Reactor although commercial fuel was stored and processed as well. The facility reclaimed uranium and plutonium from the spent fuel using a process similar to the PUREX process used at Hanford. The decontamination and decommissioning of the plutonium-contaminated facilities is in progress.

## APPENDIX C

### FACILITY DESIGN

Design criteria are established to provide technical direction and guidance for the planning and designing of new facilities and of facility additions and alterations, and the development of specifications for building acquisitions. While there is little probability that large new plutonium facilities will be constructed, there may be significant modification to existing facilities. Facility design criteria for DOE plutonium facilities can be found in DOE Order 6430.1A, General Design Criteria (DOE, 1989b). While all of the General Design Criteria manual is applicable to the design of new facilities or modification of existing facilities, Division 1300, "Special Facilities," parts 1304 and 1305, are especially pertinent to plutonium-facility design. Additionally, 10 CFR 835 Subpart K Design and Control also provides requirements for the design of radiological facilities.

This appendix provides guidance in the design of plutonium facilities. This guidance should be used to supplement the required criteria in DOE Order 6430.1A, the guidance of the DOE Radiological Control Manual (DOE, 1994l), and other DOE Orders and standards. Consideration and implementation of certain design features may be necessary to ensure meeting criteria. Other safety areas such as industrial hygiene and industrial safety are beyond the scope of this manual and are not specifically included; however, Federal and state regulations applicable to those disciplines must also be met. Further, DOE Policy 450.4 Safety Management System Policy (DOE, 1996d) provides the safety management system specifications for DOE facilities.

Radiation protection in nuclear facilities is usually achieved by a mixture of engineered and administrative safeguards. A building equipped with a maximum of engineered safeguards and a minimum of administrative controls should be more economical to operate than one with the reverse characteristics. Radiation-protection efforts may be significantly enhanced by the incorporation of the appropriate design features rather than relying on administrative controls. However, in many instances, the designer and the health physicist must balance competing objectives to attain the most cost-effective design with a high degree of safety and reliability. In designing a new facility, all of the necessary physical features can be included; however, in an old facility it may be physically or economically impossible to meet all of the requirements.

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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The guidance presented relates to physical safety and control systems only; guidance related to administrative control is not included. The phrase "safety and control systems" is used here to refer to the physical, engineered features that are used to provide radiation and contamination control. In addition to the radiation-protection requirements, facilities that contain more than 450 grams of plutonium are subject to criticality safety requirements, which include the need for a criticality alarm system and criticality dosimeters. Guidance on the security and safeguards of nuclear material (including prevention of theft or diversion) is not included but also must be considered in the design of the facility.

The following sections address the applicable standards and guides, design objectives, structural guidance, building layout, service and utility systems, and special monitoring, safety, and other systems required for the design of a plutonium facility.

## **C.1 APPLICABLE STANDARDS AND GUIDES**

The design criteria in Divisions 1304 and 1305 of DOE Order 6430.1A, General Design Criteria (DOE, 1989b), which pertain specifically to plutonium facilities, shall be applied for all new facilities that contain substantial quantities of plutonium. A facility that will handle more than 1 gram of plutonium, under certain specific conditions, shall also meet the security requirements of DOE Order 5632.1C, Protection and Control of Safeguards and Security Interests (DOE, 1994q). The design of a plutonium facility should consider the requirements and analyses described in DOE Order 5480.23, Nuclear Safety Analysis Reports (DOE, 1992d), to ensure compatibility. DOE Order 420.1 Facility Safety (DOE, 1995c) contains several requirements important to facility design. The following DOE standards may also be useful:

- DOE-STD-1024-92 Guidelines for Use of Probabilistic Seismic Hazard Curves at DOE Sites. (DOE, 1992b)
- DOE-STD-1021-93 Natural Phenomena Hazards Performance Categorization Guidelines for Structures, Systems, and Components. (DOE, 1993b)
- DOE-STD-1020-94 Natural Phenomena Hazards Design and Evaluation Criteria for Department of Energy Facilities. (DOE, 1994i)

*Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*

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- DOE-STD-1022-94 Natural Phenomena Hazards Site Characterization Criteria. (DOE, 1994r)
- DOE-STD-3009-94 Preparation Guide for U.S. Department of Energy Nonreactor Nuclear Facility Safety Analysis Reports. (DOE, 1994s)
- DOE-STD-1023-95 Natural Phenomena Hazards Assessment Criteria. (DOE, 1995e)

## C.2 DESIGN OBJECTIVES

The design of a plutonium facility shall achieve the following objectives [from DOE Order 6430.1A, General Design Criteria (DOE, 1989b)]:

- Protect the public and facility personnel from hazards associated with the use of radioactive and other hazardous materials as a result of normal operations, anticipated operational occurrences, and design-basis accident conditions, including the effects of natural phenomena pertinent to the site, and maintain these effects ALARA;
- ensure compliance with DOE policies regarding nuclear safety, criticality safety, radiation safety, explosives safety, industrial safety, fire protection, and safeguards and security protection for special nuclear material;
- protect government property and essential operations from the effects of potential accidents; and
- minimize exposures of personnel and the general public to hazardous material by emphasizing ALARA concerns during all design, construction, and operational phases.

10 CFR 835.1002 Facility design and modifications provides requirements for the design objectives.

- Optimization methods shall be used to assure that occupational exposure is maintained ALARA in developing and justifying facility design and physical controls.

- The design objective for controlling personnel exposure from external sources of radiation in areas of continuous occupational occupancy (2000 hours per year) shall be to maintain exposure levels below an average of 0.5 mrem (5 microsieverts) per hour and as far below this average as is reasonably achievable. The design objectives for exposure rates for potential exposure to a radiological worker where occupancy differs from the above shall be ALARA and shall not exceed 20 percent of the applicable standards in § 835.202.
- Regarding the control of airborne radioactive material, the design objective shall be, under normal conditions, to avoid releases to the workplace atmosphere and in any situation, to control the inhalation of such material by workers to levels that are ALARA; confinement and ventilation shall normally be used.
- The design or modification of a facility and the selection of materials shall include features that facilitate operations, maintenance, decontamination, and decommissioning.

In addition, the DOE Radiological Control Manual (DOE, 1994l) provides the following design considerations:

- Individual worker dose should be ALARA and less than 500 mrem/y;
- discharges of radioactive liquid to the environment are covered by the provisions of DOE Order 5400.5 (DOE, 1990b) and should not degrade the groundwater;
- control of contamination should be achieved by containment of radioactive material;
- efficiency of maintenance, decontamination and operations should be maximized;
- components should be selected to minimize the buildup of radioactivity;
- support facilities should be provided for donning and removal of protective clothing and for personnel monitoring, where appropriate; and
- a neutron quality factor of 20 should be used for design purposes.

The DOE Radiological Control Manual also states that facilities currently under construction should be evaluated and the above criteria applied where practicable. The manual strongly discourages locating lunch rooms or eating areas, restrooms, drinking fountains, showers and similar facilities and devices and office spaces within Radiation Areas, High and Very High Radiation Areas, Contamination and High Contamination Areas, Airborne Radioactivity Areas, Radioactive Material Areas, or and Radiological Buffer Areas.

The specific facility design chosen depends on the quantity and form of plutonium that will be used. Some simple processes involving very small quantities of unsealed plutonium can be carried out safely in well-designed and adequately filtered open-faced hoods such as those found in a general radiochemistry facility. The specific quantity that can be handled in this manner depends on the complexity of the process and the specific form of the material. Any use of unsealed plutonium should be reviewed by the facility's safety personnel, and the feasibility of the proposed use should be established based on the form of the material to be used, the work to be performed, and the engineered and administrative controls to be employed. Based on experience, if the quantity of plutonium is 100 mg or more, the process should be performed in a plutonium facility.

The application of these guidelines to specific proposals for the modification of existing facilities or the construction of new facilities requires that judgments be made based on detailed information about the facility, its use, quantities of plutonium involved, operations to be performed, degree of need for operating continuity during and/or after postulated accidents, and the potential impact on surrounding facilities and the public. The results of safety analysis reports required by DOE Order 5480.23, Nuclear Safety Analysis Reports (DOE, 1992d) will provide necessary information for design parameters. For some facility modifications, the engineering criteria outlined here may be modified or reduced if administrative requirements are increased. A cost-benefit analysis should be performed to make this decision.

The primary goal of the design objectives is to keep the plutonium confined in its intended place (i.e., capsule, hood, glove box, etc.), both during normal operations and under accident conditions. Of equal importance is consideration of the human factors in design that promote efficiency and ease of operation. Additional design criteria may be necessary in considering the requirements for decontamination, decommissioning, and dismantling (discussed in Section 10.0) of the facility when it no longer is needed.

### **C.2.1 General Design Considerations**

It is DOE's policy to design, construct and operate its facilities such that the exposure of employees and the public to hazardous materials is maintained ALARA. Detailed guidance for implementing ALARA and the application of the optimization process to facility design is provided in PNL-6577, Health Physics Manual of Good Practices for Reducing Radiation Exposure to Levels That Are As Low As Reasonably Achievable (Munson et al., 1988). Additional guidance is provided in ICRP Publication 55, Optimization and Decision Making in Radiological Protection (ICRP, 1989), and CONF-920468, Proceedings of the Department of Energy ALARA Workshop (Dionne and Baum, 1992).

Equipment reliability and human-factors engineering should be considered in the design of plutonium facilities. Both of these factors may significantly affect radiation doses and the effectiveness of personnel response to abnormal conditions. Reliability data may be available for much of the equipment that will be used. If industry information is not available, reliability analyses should be conducted. The degree of reliability that is justified may require an evaluation of the cost of the reliability versus the expected dose reduction. Human-factors engineering, as described in Part 1300-12 of DOE Order 6430.1A, General Design Criteria (DOE, 1989b), is applicable to the design of the work environment and human-machine systems at DOE facilities. Additional guidance is provided by reference in Part 1300-12.

The equipment should be designed such that the failure of a single component does not result in the loss of capability of a safety class system to accomplish its required safety function. Analyses of hazards and assessments of risks should be made during conceptual and preliminary design activities and further developed during the detailed design phase. The safety analyses should be performed in accordance with the following DOE Orders:

- DOE Order 5480.22, Technical Safety Requirements (DOE, 1992g);
- DOE Order 5480.23, Nuclear Safety Analysis Reports (DOE, 1992d); and
- DOE Order 6430.1A, General Design Criteria (DOE, 1989b).

In the planning and designing of buildings, other structures, and their operating components and systems, all aspects of operation and maintenance should be considered. This includes accessibility, dismantling, replacement, repair, frequency of preventive

maintenance, inspection requirements, personnel safety, and daily operations. Facility planning and design should use the knowledge and experience of those persons who will be responsible for operating and maintaining the completed facility. The "lessons learned" from the operation and maintenance of existing facilities should be used to avoid repeating mistakes made in past designs.

If possible, equipment that requires periodic inspection, maintenance, and testing should be located in the areas that have the lowest possible radiation and contamination levels. For equipment that is expected to be contaminated during operation, provisions should be made for both in-place maintenance and for removal to an area of low dose rate for repair. Maintenance areas for repair of contaminated equipment should include provisions for containment or confinement of radioactive materials.

Engineered safety and control systems should be designed so that they continue to function during and following an accident or emergency condition. The need for emergency systems and facilities should be determined and incorporated in the facility design. Facilities should be designed to facilitate the arrival and entry of emergency personnel and equipment in the event of a radiological emergency and to allow for access by repair/corrective action teams.

Equipment should be available to allow for an early and reliable determination of the seriousness of an accident or abnormal event. Depending upon the consequence of potential accidents, consideration should be given to relaying all such equipment alarms to a central control system or a continuously manned area. Installed on-line equipment should be protected to the extent necessary to ensure its reliability under accident conditions. To further enhance equipment reliability, the emergency equipment should, to the extent practicable, be the same equipment used for routine operations.

Emergency-power requirements that need to be satisfied and the means to provide the power should be identified in the design effort.

Emergency radiological equipment should be installed or located in areas that permit periodic inspection, testing, calibration, and maintenance.

Additional emergency-preparedness guidance is provided in Section 9.0.

## **C.2.2 Confinement**



The confinement system is a series of physical barriers that, together with a ventilation system, minimizes the potential for release of radioactive material into work areas and the environment under normal and abnormal conditions. The primary design objective for the confinement system should be to minimize exposure of the public and plant personnel to airborne contamination in accordance with DOE Order 6430.1A, General Design Criteria (DOE, 1989b). Plutonium should be separated from the ambient environment by at least two barriers and from an operator by at least one barrier.

Primary confinement refers to the barrier that is or can be directly exposed to plutonium, e.g., sealed process equipment (pipes, tanks, hoppers), glove boxes, confinement boxes, open-faced hoods, conveyors, caissons, and cells and their ventilation systems. The primary confinement barrier prevents the dispersion of plutonium through either sealed construction or atmospheric pressure differential or a combination of both. For example, process equipment that is not sealed but contains plutonium material in process should be enclosed in glove boxes or other confinement barriers. Fuel-rod cladding, bags, and other sealed containers can be considered primary confinement. The chemical reactivity and the heat-generation effect of the plutonium compound should be considered when selecting primary confinement material.

The primary confinement barrier protects operators from contamination under normal operating conditions. This type of barrier is likely to be breached under accident conditions (e.g., glove rupture, damaged seals, improper bag-out operations, or leaks of flanged joints).

The primary confinement (with the exceptions of fuel rods, sealed sources, or sealed cans) should be maintained at a negative air pressure with respect to the secondary confinement in which it is located, and it should be exhausted through a ventilation system that uses HEPA filters. The barrier and its accessory equipment should be designed to prevent accidental flooding. All primary confinement piping joints should be tested for leak tightness. Penetrations in the primary confinement barrier, such as conduit, ports, ducts, pipes, and windows, should be protected against the release of radioactive material.

Where necessary, recycle ventilation systems may be used in process enclosures, hot cells, and canyons. Inert gas systems should be designed as recycle systems, unless it is impracticable to do so. Recycled inert gas systems should be maintained completely within the primary barrier system. Extreme caution should be exercised in the use of recycle systems for contaminated or potentially contaminated air. A recirculation system should not direct air to an area where the actual or potential contamination is less than the area from which the air originated. The decision to use a recirculation system in a contaminated area should be based on a documented safety evaluation that compares the risks versus the benefits, in accordance with DOE Order 6430.1A, General Design Criteria (DOE, 1989b). Filtration should be provided to limit the concentrations of radioactive material in recirculated air to ALARA levels. The design should allow for in-place testing of HEPA filters or filter banks.

Continuous sampling and monitoring of recirculated air for airborne radioactive material should be provided downstream of fans and filters. Monitoring should be provided for the differential pressure across the filter stages and for airborne radioactive material behind the first HEPA filter or filter stage. The means for automatic or manual diversion of airflow to a once-through system or stage should be provided. The monitoring system alarm should result in the automatic diversion of airflow to a once-through system or a parallel set of filters if an automatic system is used.

The secondary confinement barrier encloses the room or compartment in which the primary confinement barrier is located, and provides contamination protection for plant personnel who are outside of the secondary confinement area. High-efficiency particulate air filtration should be required for air supplied to and exhausted from a secondary confinement barrier. Secondary confinement rooms, compartments, or cells should be separated from each other by fire doors or stops. Both the barrier walls and the fire doors should be constructed of materials that are capable of withstanding a design-basis accident. The secondary confinement should be designed for pressures that are consistent with the criteria for the ventilation system. The secondary confinement area should be at a positive air pressure with respect to the primary confinement areas and at negative pressure with respect to the outside environment and adjacent building areas that are not primary or secondary barriers.

The building is the structure that encloses both the primary and secondary confinement barriers, as well as the offices, change rooms, and other support areas that are not expected to become contaminated. It is the final barrier between the potential contamination and the outside environment. The building structure or any portion thereof may serve as the secondary confinement barrier if the requirements for both structure and confinement are met. The portion of the structure that houses activities involving radioactive material in a dispersible form should be able to withstand design basis accidents, site-related natural phenomena, and missiles without a breach of integrity that would result in releases of radioactive material from the structure in excess of DOE guidelines.

### **C.2.3 Safety Basis Analysis**

A contractor who is responsible for the design, construction, or operation of DOE nuclear facilities, as designated in writing by the Program Secretarial Officer, is required to perform a safety analysis [DOE Order 5480.23, Nuclear Safety Analysis Reports (DOE, 1992d)]. The safety analysis shall evaluate the adequacy of the safety basis for each facility. *Safety basis* is defined as the combination of information relating to the control of hazards at a nuclear facility (including design, engineering analyses, and administrative controls) upon which DOE depends for its conclusion that activities at the facility can be conducted safely. The level of safety analysis to be performed must be commensurate with the magnitude of the hazards being addressed, the complexity of the facility and/or systems being relied upon to maintain an acceptable level of risk, and the stage of the facility life cycle. The safety analysis report (SAR) defines the safety basis, document the logic of its derivation, demonstrate adherence to the safety basis, and justify its adequacy. Included in this process is evaluation of the design features and their contribution and/or mitigation to the safety basis. The SAR and the analyses performed describes the design basis accidents considered, the risk-dominated accident scenarios that have been analyzed, and the measures taken to eliminate, control, or mitigate the consequences. The design elements discussed in DOE Order 420.1, Facility Safety (DOE, 1995c), shall also be considered.

Structural design including loading combinations and construction of critical items should, as a minimum, be in accordance with current editions of pertinent nationally

recognized codes and standards. All other facility design features shall conform to applicable criteria as specified in DOE Order 6430.1A, General Design Criteria (DOE, 1989b) and to other site- or process-specific criteria developed for the facility.

### **C.3 STRUCTURAL CRITERIA**

The structure and its associated critical equipment and the ventilation, electrical, fire protection, and utility systems shall be designed to limit the release of radioactive materials during design-basis accidents postulated for the facility [DOE Order 6430.1A (DOE, 1989b)].

The structural design, the load combinations, and the construction of critical safety and fire protection features should be in accordance with the latest edition of applicable nationally recognized codes. When local codes or regulations are more stringent than the nationally recognized codes, the local codes should be followed.

#### **C.3.1 Tornado Resistance**

Critical operating areas of the facility should be designed to withstand the design-basis tornado. Specific information on site-specific tornado hazard curves, rotational speeds, elastic or plastic design methods, and other design criteria are provided in UCRL-53526 (Coats, 1984).

If site-specific hazard model studies are not available, a hazard model should be developed that is consistent with the approach used in UCRL-53526 (Coats, 1984).

#### **C.3.2 Lightning Protection**

Lightning protection should be provided for all facilities. Lightning protection systems should be designed in accordance with the most current National Fire Protection Association (NFPA) code covering lightning protection.

#### **C.3.3 Seismic Design Requirements**

The basic seismic parameters should be derived from DOE site-specific hazard model studies summarized in DOE-STD-1020-94 (DOE, 1994i). If site-specific hazard model studies are not available, a hazard model should be developed that is consistent with the approach used in DOE-STD-1020-94 (DOE, 1994i). Specific guidance on relating frequency of occurrence to facility hazard levels may be obtained from UCRL-15910 (LLNL, 1989).

#### **C.3.4 Other Natural Phenomena**

Design loads and considerations for other natural phenomena should provide a conservative margin of safety that is greater than the maximum historical levels recorded for the site. Protection against flooding should be based on no less than the probable maximum flood (PMFL) for the area as defined by the Corps of Engineers. The possibility of seismically induced damage or failure of upstream dams should be taken into account in assessing the nature of the flood protection that is required for the facility. If the facility is in a location that may be subject to ashfall from volcanic action, consideration should be given to the effects of ashfall on ventilation and electrical systems.

### **C.3.5 Explosion, Internal Pressurization, Criticality, and Other Causes of Design-Basis Accidents**

Analyses should be made to determine the probable consequences of design-basis accidents (DBAs), and critical areas and critical items should be designed to withstand DBAs. The portion of the ventilation system that is an integral part of the critical areas should be designed to withstand DBAs so that it will remain intact and continue to act as a confinement system. Building ventilation is an important part of confinement barriers and, in some cases, air flow may become the only barrier.

## **C.4 BUILDING LAYOUT**

Building layout is extremely important in the operation of a plutonium facility. Improper or poor layout can lead to operational difficulties and in some instances can contribute to the development of abnormal situations that may affect personnel safety, result in unnecessary exposure to the worker and the public, and/or increase the cost of operating the facility. Normally, three areas are involved in the overall building layout. These areas may be described as:

- The process area, where plutonium or other radioactive or hazardous materials are used, handled, or stored;
- the controlled area, which is normally free of radioactive material but could potentially become contaminated; and
- the uncontrolled area, which includes all areas where no radioactive materials are permitted and radiological controls normally are not necessary (e.g., offices and lunchrooms).

The terms *controlled area* and *uncontrolled area* defined above refer to radiologically controlled and uncontrolled areas. These terms are not to be confused with the formal definitions of controlled and uncontrolled areas related to safeguards and security concerns.

#### **C.4.1 Objectives**

The following objectives should be achieved in the design layout of the facility:

- Planned radiation exposures to personnel shall be within the prescribed limits of 10 CFR 835 Occupational Radiation Protection (DOE, 1993c). . 10 CFR 835.1002 contains requirements for ALARA design objectives for the design of new facilities or the modification of old facilities.
- The planned or unintentional release of radioactive materials from the facility should be confined to the limits of DOE Order 5400.5 (DOE, 1990b), and should be ALARA.

#### **C.4.2 General Design Criteria**

All planned processing, research and development (R&D), scrap- and waste-handling, analytical, storage, shipping, and receiving operations should be accommodated. Receiving operations that involve removal of radioactive material from protective shipping containers should be performed in a handling area that has provisions for confinement.

Real-time or near real-time accountability systems should be incorporated if possible.

The possibility of operating with multishifts per day should be taken into account in allocating space for personnel support facilities and for any special equipment that might be required to support multishift operations.

Areas should be compartmented to isolate the high-risk areas, thereby minimizing productivity and financial loss if an accident occurs.

A modular construction concept should be used where feasible to facilitate recovery from operational accidents and DBAs and to provide versatility.

All movement of personnel, material, and equipment between the process area and the uncontrolled area should be through a controlled area or an air lock. Doors that provide

direct access to the process area from the uncontrolled area (including the outside of the building) should not be permitted. If such doors are required by existing design and operating requirements for emergency exits, special administrative controls should be implemented to ensure adequate ventilation and radioactivity control. All such doors should have airtight seals. Doors without air locks should have alarms that sound when the doors are opened to signal a breach in the contamination control system.

Personnel exits should be provided in accordance with the NFPA Life Safety Code (NFPA, 1985). Personnel working in areas where an accidental breach of primary confinement will expose them to radioactive material should be located within 75 ft of an exit that leads into the next confinement barrier. Such a barrier should be a partition separating two different air-control zones, the area of refuge being on the upstream side of the barrier. The airflow through the barrier should be in the opposite direction of the exit travel.

Normal administrative traffic should be restricted to the uncontrolled and controlled areas and should not require passage through the process area. Process traffic should be restricted to process and controlled areas and should not require passage through uncontrolled areas.

Consideration should be given for provision of a ready room near or within the process area where maintenance, operating, and monitoring personnel may be readily available. The room should be in a low background area. Storage should be provided for instruments and tools needed for routine work.

Process areas should be located to permit ease of egress and material movement to ensure rapid evacuation in case of an accident and minimum potential for contamination spread during movement of material.

Indicators, auxiliary units, and equipment control components that do not have to be adjacent to operating equipment should be installed outside of radiation or contaminated areas. Units and components without internal contamination that are located in radiation areas should be designed so that they can be removed as quickly as possible.



Equipment that requires frequent servicing or maintenance should be of modular construction, standardized to the extent possible, and located outside the process area if possible.

In radiation areas, work spaces around equipment that require maintenance (e.g., pumps, valves) should be shielded to conform to the design-basis radiation levels.

Provisions should be made for the quick and easy removal of shielding and insulation that cover areas where maintenance or inspection are necessary activities. Equipment should be designed to permit visual inspection wherever possible.

Passageways should have adequate dimensions for the movement, repair, installation, or removal of proposed or anticipated equipment.

Ergonomic factors should be considered in the selection and placement of equipment components to facilitate operation and maintenance.

In any area where personnel may wear protective clothing or use breathing-air systems, the use of sharp equipment projections, which could tear clothing or breathing-air system hoses or cause wounds should be avoided.

Water-collection systems should be provided for water runoff from any controlled area. Water from firefighting activities should be considered. The collection systems should be designed to prevent nuclear criticality, to confine radioactive materials, and to facilitate sampling and volume determinations of waste liquids and solids.

Area drainage and collection systems should be designed to minimize the spread of radioactive contamination, especially to areas occupied by personnel.

Curbs should be constructed around all areas that house tanks or equipment that contains contaminated liquids to limit the potential spread of liquids, in accordance with DOE Order 6430.1A, General Design Criteria (DOE, 1989b).

Noncombustible and heat-resistant materials should be used in radiation areas that are vital to the control of radioactive materials and in equipment that is necessary for the operation of radiological safety systems. These materials should be resistant to radiation damage and should not release toxic or hazardous by-products during degradation, in accordance with IAEA Safety Series No. 30 (IAEA, 1981).

Floors, walls, and ceilings should have a smooth, impervious, and seamless finish. The junction between the floor and walls should be covered, and corners should be rounded. Light fixtures should be designed to be sealed flush with the ceiling surface to minimize horizontal surfaces and prevent entry of contamination into the fixtures in accordance with IAEA Safety Series No. 30 (IAEA, 1981). Protective coatings (e.g., paint) used in radiation areas should meet the criteria in ANSI N512-1974, Protective Coatings (Paints) for the Nuclear Industry (ANSI, 1974b).

An emergency lighting system should be provided in radiation areas to facilitate egress in emergencies. The emergency lighting should meet the requirements of the latest version of NFPA 101 (NFPA, 1985).

#### **C.4.3 Process Area**

The plutonium process area is typically a group of contiguous rooms that contain all operations involving plutonium, including processing, shipping, receiving, storage, and waste-handling. To the maximum extent practicable, the facility design should provide sufficient space and versatility to accommodate equipment for programmatic changes and process modifications.

The initial line of defense to protect workers in a process area is the primary confinement system, which includes enclosures, glove boxes, conveyor lines, the ventilation system, and process piping. The primary confinement system should be designed to minimize the impact of accidents and abnormal operations on people, facilities, and programs. The type of confinement enclosure used (e.g., hood, glove box, remote operation cell) depends on the amount and dispersibility of unsealed plutonium that will be handled and on the process involved. Generally, if the quantity of unsealed plutonium exceeds 100

mg, the use of a glove box should be considered. However, the applicability of this guideline will vary based on the individual merits of each case.

#### **C.4.3.1 Piping and Valves**

Piping and valves for radioactive liquids should not be field-run (i.e., pipe and valve locations should be located as specified on approved drawings and not at the discretion of the installer).

Notches, cracks, crevices, and/or rough surfaces that might retain radioactive materials should be avoided in the design of radioactive piping systems.

The piping system that collects contaminated liquids should be designed so that effluent from leaks in the system can be collected without releasing the liquids into personnel-access areas or to the environment.

When component or system redundancy is required, sufficient separation of equipment should be employed so that redundant systems (or equipment) cannot both be made nonfunctional by a single accident.

Stainless steel should be used in all radioactive waste and process system piping and equipment to ensure that smooth, nonporous, corrosion-resistant materials are in contact with contaminated liquids. For some applications, polyvinylidene fluoride (PVDF) piping may be preferred for inside of confinement enclosures because of its ease of fabrication, smoothness, nonporosity, and corrosion-resistance. However, it undergoes severe degradation after about  $7 \times 10^7$  rad of exposure. In general, organic materials should not be used in process-piping systems. Other materials may be used if engineering analyses demonstrate that criteria are met for strength, smoothness, porosity, and corrosion resistance for the liquids to be handled.

Piping systems used for conveying radioactive and corrosive materials should be of welded construction whenever practicable. Flanges should be used only when absolutely necessary for servicing.

Positive measures should be taken to prevent any radioactive material in the facility from entering a utility service. This may be achieved by using backflow-prevention devices and by prohibiting direct cross-connections inside the facility. The most successful backflow-prevention device is the deliberate separation of lines.

Every pipe that enters or exits a process cell or contaminated area from or to occupied areas should be equipped with block valves.

Process-piping systems carrying radioactive liquids should be designed to eliminate traps wherever possible and to permit flushing and draining except for those with loop seals. Floor drains should have the capability to be sealed.

Reduction in the size of pipelines in contaminated process-piping systems should be made with eccentric reducers installed flat-side down to avoid the formation of traps. Eccentric reducers are necessary only for horizontal pipe runs.

Changes in the direction of process piping should be made with long-radius elbows or bends. Long-radius bends should be used, where practicable, except in lines that transport solids, where blinded tees or laterals have been proven to prevent erosion. Blinded tees will encourage solids buildup. The number of bends should be minimized and pipe diameter should be increased.

If gaskets are required in process piping or associated hardware, the selected gasket material should not deform or degrade and leak when in service. Teflon should be avoided for most applications but, if needed, its use will require implementation of a most rigorous inspection routine to ensure recognition of degradation and replacement prior to failure.

Except for shielding walls, pipe sleeves should generally be provided when piping passes through masonry or concrete walls, floors, and roofs. The sleeves should be sloped to drain toward the controlled area. The space between the pipe and the sleeve should be packed and sealed. If the sleeve is to be sealed, then additional provisions should be made for draining the annulus.

If underground piping for transporting radioactive or hazardous materials is required, it should be installed inside another pipe or tunnel that provides a second barrier to the soil. Provisions to detect a failure in the primary piping (leak detection) should be provided. An effective solution may be to install a double-walled pipe with an annular space that can be sampled at intervals not exceeding 300 feet. The underground piping should also have cathodic protection.

All valves that are not functionally required to be in contact with contaminated liquids should be located in nonradiation areas (e.g., steam, air, water) in accordance with IAEA Safety Series No. 30 (IAEA, 1981).

Process valves should not be located at low points in the piping except in cases where it is necessary in order to properly drain the piping when needed.

Valve seals and gaskets should be resistant to radiation damage.

Straight-through valves generally should be used to simplify maintenance and minimize particle traps.

Valves should be designed to operate in the stem-up orientation, which would limit potential leakage when the pipe is unpressurized. Valves and flanges should be located to minimize the consequences of contamination from leaks.

Generally, process solutions should have primary and secondary confinement. However, in rare instances where process solutions are allowed to flow outside of confinement, they should flow only by gravity and the pressure head should be limited to an equivalent of about 10 feet of water.

The corrosion resistance of the primary block valve and/or check valve and all associated piping in the in-cell and/or contaminated areas should be equivalent.

The use of pumps in contaminated piping systems should be avoided to reduce potential contamination problems that result from pressurization and to reduce

the maintenance requirements associated with pumping. The use of gravity flow, jets, vacuum, or airlifts is a suitable alternative. Vacuum transfers are preferred. If jets or airlift transfers are used, an adequate waste-air cleanup system should be provided.

#### **C.4.3.2 Structure**

Floors should be designed in accordance with code requirements considering the maximum loads anticipated.

#### **C.4.3.3 Storage**

In-process storage should not be permitted; however, temporary storage of the product in the process area until it can be taken to an appropriate storage area should be permitted.

Storage facilities in the process areas should be designed to prevent the exposure of operating personnel and to meet the requirements for security and safeguards as given in DOE Order 5632.1C, Protection and Control of Safeguards and Security Interests (DOE, 1994q), and other DOE Orders in the 5630 series that comprises the DOE safeguards program to guard against theft or unauthorized diversion of special nuclear material.

#### C.4.3.4 Shielding

Provisions should be made to accommodate the shielding of all items in the process area. All structures (e.g., floors, walls, and glove boxes) may require additional shielding during the lifetime of the facility because of increased throughput or higher radiation levels of the material being processed.

DOE Order 6430.1A, General Design Criteria (DOE, 1989b) establishes a radiation level of 1 rem/y to the whole body as a design guide. However, the DOE Radiological Control Manual (DOE, 1994l), Article 128, "Facility Modification and Radiological Design Considerations," states that "individual worker dose should be ALARA and less than 500 mrem per year." The design of a routinely occupied portion of a process area should never be based on anticipated dose rates in excess of 100 mrem/h.

Concrete radiation shielding should be in accordance with ANSI N101.6-1972, Concrete Radiation Shields (ANSI, 1972).

Straight-line penetration of shield walls should be avoided in order to prevent radiation streaming.

Robotics and/or shielded operations performed remotely should be used as much as practicable and should be used where it is anticipated that exposures to hands and forearms would otherwise approach 10 rem/y. Also, robotics or other non-hand contact methods should be used where contaminated puncture wounds could occur.

Shielding materials should be noncombustible or fire resistant, to the maximum extent practicable.

**C.4.3.5 Confinement Devices**

Different devices may be used to confine and control radioactive material. The selection of the appropriate device will depend on the quantity of material, its form, and the operations to be performed. For specific operations, encapsulation may be the confinement of choice. Sealed source containers should be designed to prevent contact with and dispersion of the radioactive material under all normal conditions and when inadvertently dropped. Sealed sources should be shielded as required to ensure that personnel in routinely occupied areas do not receive more than 0.5 mrem/h.

Seismic protection should be provided to minimize movement of confinement enclosures if ground movement occurs.

Fume hoods may be used for some operations with plutonium, depending on the quantity and dispersibility of the material. In general, plutonium fume-hood operations should be limited to wet chemistry processes and less than 100 mg of plutonium. For some operations, such as metallography and x-ray analysis, larger quantities may be handled. The location of each hood should be evaluated with respect to ventilation supply and exhaust points, room entrances and exits, and normal traffic patterns. Hood faces should not be located within 10 ft of the closest air supply or room exhaust point, which might disturb airflow into the hood. Hoods should not be located in or along normal traffic routes.

An open-faced hood should be designed and located to provide a constant air velocity across the working face. A face velocity of greater than 125 linear ft/min over the hood face area should be provided to ensure control of radioactive materials. Much of the nuclear industry uses 150 linear ft/min as the criterion. If room air currents might upset the uniform entrance of air, the hood-exhaust requirements should be increased. Turbulence studies may be necessary to verify adequate control of radioactive material. Physical stops should be provided to ensure that the required hood face velocity is maintained.



Hood design and filtration systems should comply with the criteria established in ERDA 76-21, Nuclear Air Cleaning Handbook (ERDA, 1976); Industrial Ventilation, A Manual of Recommended Practice by the American Conference of Governmental Industrial Hygienists (ACGIH, 1988); and ORNL-NSIC-65, Design, Construction, and Testing of High Efficiency Air Filtration Systems for Nuclear Applications (ORNL, 1970).

The hood structure should have a smooth, corrosion-resistant inner surface that is made of or coated with easily cleaned material.

Glove boxes, conveyors, and other enclosures should be designed to control and minimize the release of radioactive materials during normal operations and postulated accidents. Noncombustible or fire- and corrosion-resistant materials should be used in the construction of the confinement system, including any shielding employed. Fixed modular construction should be employed wherever possible, using a standardized attachment system that will permit replacement or relocation of the contents within the glove-box system with a minimum spread of contamination. Fire dampers should be provided between glove boxes to limit the spread of fire. Fire dampers should be tested frequently to assure proper operation when needed.

The process design should minimize required hands-on operation in glove boxes and other primary confinement units.

The glove-box design should include sufficient work space to permit removal of materials and easy personnel access to all normal work areas, and it should provide for the collection, packaging, storage, and/or disposal of waste generated by the operation of the glove box.

Consideration should be given to incorporating transfer systems such as a double-door sealed transfer system for removal of plutonium from a glove box. Types of removal and transfer systems are given in IAEA Safety Series No. 30 (IAEA, 1981). These types of removal systems are designed to permit entry and removal of material without breaching the integrity of the glove box.

Automatic glove-hanging systems are available and should be considered in the design or modification of glove box systems.

The ease of visibility for activities, accessibility of necessary in-box controls, and ease of cleanup and waste removal should be considered in the design of glove boxes. Glove boxes should be designed and constructed to reduce points of material accumulation.

Equipment should be designed to preclude sharp corners, barbs or pointed parts, and pinching points that could puncture glove box gloves or skin. All corners should be rounded and burrs removed.

Ergonomics should be considered in designing the height of glove-box ports and access to inner surfaces and equipment.

Each glove box should be equipped with an audible alarm that can be tripped to signal radiological problems. Individuals should be able to activate the alarm without removing their arms from the glove box. The alarm should sound in a continuously occupied area where it should, as a minimum, identify the room in which the alarm originated.

A HEPA filter should be installed on the air inlet to the glove box if required to prevent the backflow of contamination. Prefilters should be installed upstream of the HEPA filter where appropriate. The exhaust outlet for each glove box should have HEPA filters to keep the ventilation ductwork clean. This filter should not be counted as a formal HEPA stage and need not meet all the test capabilities for HEPA filtration; however, it should be tested prior to installation. Push-through filter change-out systems should be used if possible. The HEPA filters downstream of the glove box should be readily accessible for filter change-out and testable.

Glove-box air inlets and inlet filters should be protected or oriented to prevent inadvertent entry of water into the box (e.g., a fire-sprinkler system discharge or water-line leak).

Glove boxes should be designed to operate at a negative pressure ( $0.75 \pm 0.25$  in. water gage (WG)) with respect to the room in which they are operated. Differential pressure gauges should be installed on each glove box or integrally connected series of glove boxes. During abnormal conditions, control devices to prevent excessive pressure or vacuum should be either positive-acting or automatic or both. The ventilation system should be designed to provide and maintain the design negative pressure during normal operations and the design flow through a breach. There should be exhaust capacity on demand that will promptly cause an inflow of air greater than 125 linear ft/min through a breach of at least a single glove-box penetration of the largest size possible. Filters, scrubbers, demisters, and other air-cleaning devices should be provided to reduce the quantities of toxic or noxious gases and airborne particulates that enter the ventilation system prior to its entry into the exhaust system.

Each glove box or integrally connected series of glove boxes should be equipped with an audible alarm that alerts personnel when a system pressure or vacuum loss is occurring. The alarm should be set at -0.5-in. WG relative to the room in which the glove box is located.

The number of penetrations for glove-box services should be minimized. The fittings should provide a positive seal to prevent the migration of radioactive material. For the same reason, penetrations for rotating shafts should not be permitted except where rotating shafts have seals. Seals for rotating shafts are very reliable and are preferred to motors inside the glove box.

Vacuum systems connected to a glove-box should be designed to prevent an evacuation and possible implosion of the glove box.

Any gas-supply system connected directly to a glove box should be designed to prevent pressurization, flow in excess of the exhaust capacity, and backflow. Flammable or combustible gases should not be used in glove boxes but, if required, should be supplied from the smallest practical size of cylinders. Flammable gas piped to a plutonium processing building should not enter the building at a pressure exceeding 6-in. water (DOE Order 6430.1A, General

Design Criteria (DOE, 1989b). Vacuum pump exhaust should be filtered and exhausted to the glove box or other acceptable exhaust system.

If process water is provided to a glove box and the water must be valved on when the box is unattended, a system should be installed to automatically close a block valve in the water-supply line if a buildup of water is detected on the box floor or in the box sump.

Process piping to and from glove boxes should be equipped with backflow prevention devices and should be of welded stainless steel construction. Vacuum breaker-type devices are generally more reliable than other types.

Glove-box components, including windows, gloves, and sealants, should be of materials that resist deterioration by chemicals and radiation.

Glove ports should be designed to allow for the replacement of gloves while maintaining control of radioactive material. The ports should be located to facilitate both operating and maintenance work. The need for two-handed operation, depth of reach, mechanical strength, and positioning with respect to other ports should be considered in the design. Covers or plugs should be provided for each port. The covers or plugs should provide shielding equivalent to the glove-box walls. Automatic glove-changing systems should be considered.

Bag-out ports, sphincter seals, and air locks should be designed and installed to facilitate the introduction and removal of equipment and supplies without compromising contamination control. Air-lock gaskets at the bottom rim should be protected from any physical damage potentially incurred by removing items. Air locks should be designed to be at negative pressure with respect to the workstation and positive pressure with respect to the glove box.

Windows should be constructed of noncombustible or fire-resistant materials that resist scratching, breaking, and radiation degradation. Wire glass should be considered except where precluded by requirements for visual acuity. In those instances, tempered or safety glass may be suitable. Windows should be kept as

small as possible while still meeting visual requirements. A push-in window design should be considered for ease of replacement. Use of PVDF lining or laminations on windows may reduce their degradation and increase the ease of their decontamination. The windows should be securely fastened and gasketed or sealed. The gasketing material should be resistant to degradation by radiation or other materials to which it will be exposed. Lighting fixtures should be mounted on the glove-box exterior to the extent practicable.

Generally, organic (plastic) materials are not recommended for use in plutonium glove boxes. However, when dealing with process streams containing large quantities of fluorides or chloride ions, organic (plastic) pipe and equipment are sometimes required. When using organics in the glove box, care must be exercised in the selection of the material to minimize alpha radiation deterioration.

Fire protection should be provided in the glove box, enclosure, and conveyor systems to meet DOE improved-risk objectives. Automatic fire suppression should be provided in all new structures over 5000 square feet and in all structures having a maximum possible fire loss (MPFL) in excess of \$1 million or where the maximum credible fire will result in the loss of use of a vital structure for a period longer than that specified as acceptable by the applicable PSO [DOE Order 420.1, Facility Safety (DOE, 1995c)]. When the MPFL exceeds \$50 million, a redundant fire-protection system should be provided that, despite the failure of the primary fire-protection system, will limit the loss to \$50 million.

Discrete workstations within an enclosure should be separated from each other by fire stops to prevent the spread of fire. Fire stops should be designed to be normally closed. For systems in which fire stops must normally be open, closure should be automatic upon actuation of the fire-sensing system. Instead of a fire-sensing system, an oxygen-deficient atmosphere may be provided as the normal or required operating atmosphere within the enclosure. Where automatic fire-suppression systems are not required, a fire-detection system should be

installed. Provisions should also be made for manual fire suppression where it is deemed necessary.

The actual sources inside the glove box should be shielded, if possible, instead of shielding the glove box. However, the glove box should be equipped with or capable of accepting any necessary neutron and/or gamma shielding.

#### **C.4.4 Controlled Area**

All support facilities that have a potential for periodic low-level contamination should be located in the controlled area. These facilities include change rooms and decontamination rooms for personnel; health physics laboratories; facilities for the receipt, temporary storage, and shipment of radioactive and potentially contaminated materials; maintenance rooms for regulated equipment; mechanical equipment rooms; and other laboratory facilities.

Air locks between controlled and uncontrolled areas should be used to provide confinement of the controlled area if an inadvertent release of radioactive materials or a fire occurs. Air locks should also be provided in controlled areas where there is a potential for radioactive contamination to be spread from an area of high contamination to one of lower contamination.

Where possible, each controlled area should have a single access and exit point for personnel during normal operation. Access points should be accessible through change rooms. Other access and exit points should be available as required for emergencies and in compliance with the NFPA Life Safety Code (most current version) (NFPA, 1985).

Space for step-off pads and radiation monitoring and survey equipment should be provided at the exit from controlled areas that are potentially contaminated and between high- and low-level contamination areas. The space provided should be sized to accommodate the expected work force.

**C.4.4.1 Change Rooms**

Change rooms should be available for both men and women, with lockers to support the anticipated number of workers and support personnel. Change rooms should include facilities for storing and dispensing clean protective clothing, a well-defined ventilated area near the exit from the controlled area for the temporary storage of potentially contaminated clothing, and adequate shower facilities. The clean side of the change room should be easily separable from the potentially contaminated side of the room.

Space for step-off pads and radiation-monitoring survey equipment should be provided for personnel and equipment leaving the controlled portion of the change room.

Liquid wastes from potentially contaminated showers should be routed to the liquid radioactive waste system or to a holding tank that may be sampled before the waste is released.

The ventilation system should be designed to prevent the spread of contamination from the controlled to the uncontrolled portion of the room.

**C.4.4.2 Personnel-Decontamination Room**

A personnel-decontamination room (or station) should be provided for each plutonium facility. It should be located near or in the change rooms. A decontamination room with the capability to decontaminate male and female personnel simultaneously should be considered. The use of installed partitions or curtains should be considered for this purpose. An adequately equipped decontamination room should have communications equipment, a workbench with a cabinet for decontamination supplies, an examination chair, a sink, and showers. Both the sink and showers should be connected to a holding tank for sampling or routed to the process waste. The room should contain equipment for performing nasal irrigations and initial surveys of nasal swipes.

#### **C.4.4.3 Health Physics Laboratory Office**

Health physics personnel in a plutonium facility should be assigned laboratory office space at or near the exit from the process area into the controlled area. As a rule of thumb for determining space needs, one radiation protection technologist should be available for every 10 radiation workers. Space should be included for the readout of radiation-protection instrumentation, preparation of radiation protection records, counting equipment, and storage of portable instruments.

#### **C.4.4.4 Mechanical Equipment**

Where possible, mechanical equipment (e.g., motors, pumps, and valves that may be a source of radioactive contamination) should be located in the process area. Enclosures that will contain the contamination should be placed around the equipment. Such enclosures should be easy to decontaminate.

### **C.5 SERVICE AND UTILITY SYSTEMS**

Utility services should be designed to provide reliability that is consistent with 1) the operational requirements for the control and confinement of radioactive materials and 2) the potential hazards under all probable conditions. The services and utilities that are important to the continuity of essential plant functions should be designed to the same integrity level as the function they serve. Some service or utility systems are connected to other systems or structures that are essential to prevent the release of radioactive materials. Such service or utility systems must be designed so that if they fail, connecting systems will remain functional.

#### **C.5.1 Ventilation Systems**

Ventilation systems include the supply and exhaust systems and the associated ductwork, fans, air cleaning, tempering, or humidity control devices, and associated monitoring instrumentation and controls required to confine radioactive materials within the



ventilation system. The design of ventilation system components does not include process vessels, primary confinement or containment housing, or the building structure.

#### **C.5.1.1 Design Objectives**

The ventilation system should be designed to confine dispersible radioactive material within prescribed areas of the facility. It should also be designed to limit airborne concentrations of radioactive material in occupied areas of the facility and in effluents to levels that are less than the applicable concentration guides and ALARA.

The ventilation system, which serves as an engineered safety and control system, should be designed to remain operational or fail safely under all operational and credible accident conditions. The failure of any single component should not compromise the ability of the system to maintain confinement of radioactive materials or control their release to the environment. Specific response requirements of the system and its components should be identified through a safety analysis.

#### **C.5.1.2 Air Flow and Balance**

The design of ventilation systems should ensure that, under all normal conditions, the air flows toward areas of progressively higher radioactive material inventory. Air-handling equipment should be sized conservatively enough that minor fluctuations in air flow balance (e.g., improper use of an air lock, or occurrence of a credible breach in a confinement barrier) do not result in air flowing from higher to lower radioactive material inventory areas. To prevent the movement of contamination from high radioactive material areas to low radioactive material areas in case of a flow reversal, HEPA filters should be provided at ventilation inlets in confinement area barriers.

A minimum of two negative-pressure zones should exist within a process building. The first, the process confinement system should serve the spaces

within the glove boxes, conveyors, transfer boxes, and other spaces that may contain plutonium during the course of normal operations. The second should serve the process areas and other potentially contaminated areas adjacent to the process-confinement system. Controlled areas that are contiguous to process areas and potentially free of contamination constitute a third zone. Some facilities have a minimum of three zones and frequently four.

A minimum pressure differential of between 0.75-in. and 1.0-in. (1.9-cm and 2.5-cm) WG, negative with respect to the room, should be maintained in all process confinement systems. A negative pressure differential of at least 0.1-in. (0.25-cm) WG should be maintained between process and controlled areas and between controlled areas and uncontrolled areas. Air locks between zones should be provided where necessary to ensure that proper differential pressures are maintained. Differential pressure between the containment enclosure and the outside atmospheric pressure may be as great as 3 in. of water [ERDA 76-21, Nuclear Air Cleaning Handbook (ERDA, 1976)].

The design of the ventilation system should include an analysis to demonstrate that the system is capable of operating under the safety-basis conditions. To the maximum extent practicable, the system should be designed to ensure that the products of combustion are not spread beyond the room of origin unless directed through appropriate ventilation channels. The exhaust system should be designed to provide cleanup of radioactive material and noxious chemicals from the discharge air and to safely handle the products of combustion.

Provisions should be made for independent shutdown of ventilation systems where this could be an advantage to operations, maintenance, or emergency procedures such as firefighting. In assessing the desirability of providing for shutdown of a ventilation system under such conditions, full consideration should be given to all possible effects of the shutdown on air flows in other interfacing ventilation systems. It may be more appropriate to provide for drastically reduced flow rather than for system shutdown. For example, reducing air supply to 10% and exhaust flow to 20% of operating values would minimize ventilation and maintain negative pressure. Positive means should be provided for

controlling the backflow of air, which might transport contamination. The ventilation system and the associated fire-suppression system should be designed for fail-safe operation.

The ventilation system should be appropriately instrumented and alarmed, with readouts in continuously occupied control rooms. A listing and the function of required and recommended instrumentation are given in ANSI N509-1989, Table 4-1 (ANSI, 1989b).

Building penetrations for ventilation ducts should be kept to a minimum and should be designed to protect the critical systems against postulated accidents. No penetrations should be permitted if the barrier around the process area is the outside wall of the building.

Room air in controlled and process areas may be recirculated if the recirculating air system is provided with two HEPA filter banks in series. One of the filter banks should be in the exhaust duct leading from the rooms where airborne activity might be introduced. An air monitor should be located between the two filters and set to alarm when the air concentration reaches a preset point. Airflow should then be diverted either manually or automatically to a once-through system using the air-monitor alarm indication to trip the system. Recirculation from a zone of higher contamination to a zone of lower contamination should be prohibited.

### **C.5.1.3 Air Supply**

Supply air should be appropriately filtered and conditioned in accordance with operational requirements and with the levels recommended for comfort.

The ventilation rate in process areas where uncontained radioactive materials are handled should be from 12 to 60 air changes per hour (ORNL, 1970) depending on whether the area is normally occupied by workers, the need for removal of process or decay heat, and the need for removal of decay fumes. A minimum of eight air changes per hour should be provided in support facilities within the process area. Adequate air filters should be used at the intake of the ventilation supply system to minimize dust in the process area and to reduce the dust loading on HEPA filters.

A downward air-flow pattern should be provided at worker locations to direct air from any potential leak point down and away from the worker's face.

Consideration should be given to the distribution of inlet air through a number of small ports or by slot-type distributors to decrease the possible occurrence of “dead spots” with little air circulation.

Glove boxes, conveyors, and other systems that require a controlled atmosphere may be equipped with a recirculating air system. All parts of the system should operate at air pressures that are negative with respect to the room. Process enclosures that use normal air may receive their air supply from the room through dust-stop and HEPA filters mounted on the glove box.

Consideration should be given to isolating process rooms from each other during accidents. The principle of compartmentation and separation should be extended to systems handling ventilation in working areas by the most practicable use of individual ventilation systems. Emergency back-up should be provided through combinations of manifolds and damper cutovers between adjacent individual ventilation systems. Redundancy can be minimized by the provision of a back-up unit for each two individual systems.

#### **C.5.1.4 Exhaust Systems**

The number of required exhaust-filtration stages from any area of the facility should be determined by analysis to limit quantities and concentrations of airborne radioactive or toxic material released to the environment during normal and accident conditions. Materials released should be in conformance with applicable standards, policies, and guidelines. In general, each exhaust filter system should consist of a minimum of two HEPA filters for room air and three HEPA filters in series for glove-box or hood-exhaust air. Only two stages of glove-box or hood-exhaust filters need to be equipped for in-place testing.

The filtration system should be designed to allow for reliable in-place testing of the HEPA filters and ease of filter replacement to the extent practicable.

The exhaust system for a glove box or hood should be separate from the exhaust system for room air. The hood exhaust system need not be separate from room-exhaust ventilation if ventilation is once-through. Exhaust air should be drawn through a HEPA filter at the glove-box or hood-exhaust point to maintain primary control at that point and minimize contamination of ductwork. This filter should not be counted as a confinement barrier unless it is testable in place. Additional HEPA filters in series should be separated at a sufficient distance to permit in-place testing of each stage of the filters.

Dampers should be installed in the glove-box, hood, and room-exhaust ducts so that required air-pressure differentials can be maintained. Automatic backflow dampers should be installed in series with the exhaust dampers. Manual controls, or automatic controls with manual override, should be provided as needed for ventilation systems or their components for flexibility of operation.

Integral fire-suppression equipment should be provided as needed within each ventilation system to ensure that a safety-basis fire could not degrade the integrity of the high-efficiency air cleaning system. Where appropriate, a cool-down chamber with water sprinklers, a prefilter demister, and a spark arrestor screen should precede the first stage of the final HEPA filtration system.

The water spray from a cool-down chamber should be automatically actuated by appropriate temperature- and smoke-sensing devices as determined by the accident analysis.

All potentially contaminated air should be exhausted through a common stack. Continuous monitoring and a representative, redundant sampling capability should be provided on exhaust stacks that may contain radioactive or toxic materials. The ventilation exhaust stack should be located as far away from any air intake as is reasonably possible. Design criteria for effluent monitoring and sampling and elements for consideration in effluent radioactivity measurement are described in DOE/EP-0096, A Guide for Effluent Radiological Measurements at DOE Installations (DOE, 1983). ANSI N42.18 provides specifications and performance of onsite instrumentation for continuously monitoring radioactivity in effluent (ANSI, 1974a).

#### **C.5.1.5 System Testing and Control**

The ventilation system is considered an essential safety and control system and should be designed in accordance with ANSI/ASME N509-1989, Nuclear Power Plant Air Cleaning Units and Components (ANSI, 1989b). The minimum acceptable response requirements for the ventilation system, its components, instruments, and controls, should be established based on results of safety analyses for normal, abnormal, and accident conditions. These requirements should include system and component design characteristics, such as the installation of standby spare units, provision of emergency power for fans, installation of tornado dampers, seismic qualification of filter units, and fail-safe valve positioners.

The ventilation system should be designed to operate effectively and to permit servicing or filter replacement while operating. The system's effectiveness should be assessable during operation by means of installed testing and measurement devices.

Air-cleaning systems should be designed for the convenient, repetitive, and reliable in-place testing of each stage of the system for which credit is taken in accordance with ANSI/ASME N510-1989, Testing of Nuclear Air Cleaning Systems (ANSI, 1989d). Provisions for in-place testing should include aerosol injection ports, sampling ports, and connecting and bypass ductwork. Each filter bank should be tested upon installation, periodically thereafter, and anytime when conditions have developed that may have damaged the filter, e.g., pressure drop, over-pressure, water spray, etc. The filter or filter bank should be tested and demonstrate a particle-removal efficiency as described in ANSI/ASME N510-1989, Testing of Nuclear Air Treatment Systems (ANSI, 1989d), and ANSI/UL 586-1990, High-Efficiency Particulate Air Filter Units (ANSI, 1990).

The portions of the ventilation system that are essential to preventing releases of radioactive materials should continue to function (or automatically change to a safe-failure mode) under abnormal or accident conditions. The ventilation system fans should produce a maximum exhaust rate that is greater than the maximum supply rate. Exhaust fans should be provided with emergency power in the event of loss of normal electrical power supplies. Exhaust and supply fans should be redundant. If the system fails, exhaust-control dampers should fail in the open position and the supply-control dampers should fail in their preset closed position. Supply fans should automatically cut off when the exhaust-fan capacity in service is not sufficient to maintain the proper pressure differential. Alarms should be provided to signal the loss of fan capacity or improper air balance. System components or devices that must function under emergency conditions should be able to be tested periodically, preferably without interruption of operations.

Appropriate surveillance instrumentation and manual system operation controls should be provided at one common location. In addition, surveillance instrumentation should be located in an external or protected area that would be accessible during and after all types of postulated accidents.

## **C.5.2 Electrical Power**

Both normal and emergency power supplies should be available to a plutonium facility to ensure that critical systems can continue to operate under both normal and accident conditions.

### **C.5.2.1 Normal Power**

A plutonium facility's normal electrical power needs should be met by two primary feeders. The preferred primary feeder should provide basic service to the facility and consist of a radial feeder connected directly to the main substation serving the area. To minimize power outages, this feeder should be an express feeder and should not have any other loads connected to it.

The alternate primary feeder should be in ready standby to provide backup power to the preferred primary feeder power supply. In the event of a forced outage or planned maintenance of the preferred primary feeder, the power load should automatically transfer to the alternate feeder. The alternate primary feeder should also be a radial feeder connected directly to a substation and should have no other loads connected to it. To minimize simultaneous outages of the preferred and alternate primary feeders due to lightning or other physical damage, the two feeders should have maximum physical separation.

### **C.5.2.2 Emergency Power**

The facility should be provided with a reliable, local source of emergency power if both primary sources fail. The emergency power source should be completely independent of the preferred and alternate primary feeders. The emergency power should be generated onsite by turbines or diesel generators with automatic starting and switch-over equipment. The emergency system should be physically separated from the normal power systems, except at the automatic transfer switch, so that any electrical or mechanical breakdown of the normal power system will not render the emergency system inoperative.



The time lag between electrical power failure and the resumption of emergency power should not exceed 20 seconds, and the emergency system should remain energized for at least 5 minutes after the restoration of primary power to allow for an orderly transition. The emergency power sources should have sufficient capacity and sufficient fuel supplies stored onsite to maintain the integrity of all critical building systems for approximately 48 hours. The amount of time that emergency power is necessary should be determined by the requirements for bringing the processes to safe shutdown condition. Chemical and thermal inertia also should be considered. The emergency power system should be able to carry identified critical loads such as air exhaust and supply systems, fire-detection and fire-suppression systems, related instrumentation and control functions, necessary criticality and radiation-monitoring instrumentation, certain processing equipment, and any other essential building systems identified during safety analysis. Sensitive safety equipment should be tested to verify that it will remain operable during the switch-over and after enduring the electrical transient.

Noncritical uses of emergency power should be avoided.

### **C.5.3 Water Supply**

Water-storage tanks with multiple or backup supplies should be provided to simultaneously supply water for fire protection, processing, and drinking.

The design of the water-supply system shall provide water for firefighting and automatic sprinkler systems in accordance with the DOE Order 420.1 (DOE, 1995c) and Factory Mutual and National Fire Protection Association Standards. The fire-protection water supply and distribution design required for critical item protection should ensure the continuity of protection in the event of postulated accidents.

Potable water should be distributed to drinking fountains, eyewash fountains, showers, emergency showers, lavatories, and noncontaminated laboratories. Raw water may be used in toilets and urinals. The potable water system shall be protected against contamination, in accordance with Division 2 of DOE Order 6430.1A, General Design Criteria (DOE, 1989b) and 40 CFR 141 and 142 (EPA, 1992a). Water mains should not

pass through process or controlled areas. Branch lines may be permitted in process areas for safety showers and fire-protection sprinkler systems only.

The facility water system preferably should be isolated from primary water mains by an air gap to prevent any possibility of contamination of public water supplies. If an air gap is not possible, reduced-pressure type of backflow prevention devices meeting the requirements of the American Water Works Association C506-78-1983 (AWWA, 1983) should be used. Process water supplied to the process and controlled areas must be isolated from the potable water system. Cross-connections should not be permitted.

#### **C.5.4 Fire Protection**

Each area in the plant building shall be equipped with fire-detection devices that are best suited for that area, as described in Division 15 of DOE Order 6430.1A, General Design Criteria (DOE, 1989b) and in NFPA National Fire Codes 71 and 72A through 72D (NFPA latest revisions) (NFPA, 1985). All equipment should be approved by a recognized testing laboratory. The spacing, sensitivity, and location of the detectors should be given careful consideration to ensure rapid response.

All fire detectors and/or automatic fire-suppression systems should be connected to fire-alarm annunciators. The annunciator system should be sufficiently subdivided to identify the location of a fire.

Fuels and combustible materials should be stored at a central facility that is remote from the plutonium-processing building(s). Piped natural gas should not be provided to plutonium process or storage areas. Separate bottled gas systems should be provided where required.

The ventilation system of the facility should be designed to withstand any credible fire or explosion. It should be constructed of noncombustible materials and have fire-detection and fire-suppression equipment, including heat and smoke detectors, alarms, fire doors and dampers, and heat-removal systems. The final filter bank of the building's air-exhaust system should be protected from damage by hot gases, burning debris, or

fire-suppression agents that may be carried through the exhaust ducts during a 4-hour fire.

Over-pressure protection should be considered for critical items such as glove boxes, cells, and ventilation ducts.

### **C.5.5 Waste**

Waste from plutonium-handling facilities includes radioactive, radioactive mixed, and hazardous (nonradioactive) materials and will be in the form of liquid or gaseous effluent and solids packaged for shipment offsite. A principal design objective for the process systems should be to minimize the production of wastes at the source. A principal design objective for the waste management systems should be to provide facilities and equipment to handle the wastes safely and effectively. The design of the facility should limit the environmental release of radioactive, radioactive mixed, and hazardous materials to less than the DOE and EPA regulations and ALARA. Emphasis should be placed on reducing total quantities of effluent (both radioactive and nonradioactive) released to the environment. See Section 8.0 for additional information on waste management.

## **C.6 SPECIAL SYSTEMS AND EQUIPMENT**

Special systems and equipment should be incorporated in plutonium facilities to ensure the safety of the worker and protection of the public. As a minimum, the following systems should be included:

- Air sampling and monitoring;
- breathing air;
- personnel monitoring;
- criticality safety;
- nuclear accident dosimeters; and
- monitoring and alarms.

These systems and equipment plus some that may not be directly related to personnel safety and radiation protection are discussed in the following subsections.

### **C.6.1 Air Sampling and Monitoring**

The air-monitoring and air-sampling systems for a plutonium facility should meet the criteria established in the Implementation Guide. Workplace Air Monitoring (DOE, 1994g). In addition, PNL-6612, The Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) provides information for the design, implementation, operation, documentation, and evaluation of a plutonium air-sampling program.

### **C.6.2 Breathing Air**

For facility design, confinement of airborne radioactive materials should be the required method of preventing internal deposition of radioactive particulates. However, during operation and maintenance of the facility, situations may occur (accidents, special maintenance, spill recovery, etc.) for which air-supplied respiratory protection is required.

A plutonium facility should be provided with a system that is capable of supplying breathing air to a number of workstations in each occupied area where the following conditions exist:

- Gaseous or airborne radioactive material may cause occupational exposure limits of 10 CFR 835 to be exceeded;
- potentially dispersible plutonium compounds exceeding 100 mg are handled outside of containment devices; or
- personnel may be required to enter cells or other areas that contain large amounts of loose radioactive material for repair, maintenance, decontamination, or operation.

Breathing-air systems may be portable or semiportable bottled systems or installed compressor systems. The facility design requirements should be determined by the system selected.

Breathing-air supply systems should meet the requirements of ANSI Z88.2-1980, Practices for Respiratory Protection (ANSI, 1980b) and 29 CFR 1910 (most recent revision) (DOL, 1993). Air-line connections for the breathing air must be unique to preclude connecting other gas supplies to the breathing-air lines. Additional criteria for design of breathing-air systems found in the references mentioned above should be considered.

### **C.6.3 Personnel Monitoring**

The facility design should provide for location of personnel monitoring devices in the vicinity of the workplace. To minimize the potential spread of radioactive contamination, personnel survey instruments should be available at suitable locations within the process area, such as for personnel exiting from glove boxes, at bag-out stations, and at exits from compartmentalized facilities. Survey instruments or monitoring instruments should be available at contamination-control change rooms and at exits from controlled areas.

### **C.6.4 Criticality Safety**

See Section 7.0 for detailed guidance on criticality safety in a plutonium facility including criticality alarms and nuclear accident dosimeters.

### **C.6.5 Other Systems**

Many systems employed within a plutonium facility are not directly related to personnel safety and radiation protection. However, because of the special impact that these systems may have on a plutonium facility, individuals responsible for personnel protection should be aware of them. Some examples are as follows:

- Process instrumentation and control indicators to monitor and maintain control over the process and to detect and indicate abnormal and accident conditions;

- surveillance systems to ensure the integrity of all process piping, tanks, and other containment equipment, including those used for liquid effluent; and
- vacuum, airlift, or gravity systems to transfer toxic or corrosive liquids or slurries.

Special controls should be provided for flammable, toxic, and explosive gases, chemicals, and materials that are used in plutonium-handling areas. Gas and chemical storage facilities, including distribution piping systems, should conform to good design practice and applicable codes and standards. Consideration should be given to compatible groupings that, under accident or leakage conditions, would minimize any adverse combining of materials. Means for remote shutoff of piping should be provided. In addition, the following rules should be observed:

- Nonflammable hydraulic and lubricating fluids should be used in the plutonium-handling area;
- protective barriers should be provided around high-pressure or other potentially dangerous systems;
- incompatible chemicals, materials, and processes should be isolated from one another; and
- pressurized gas lines used in the plutonium-handling areas must be properly vented.

Facilities for equipment maintenance should be provided.

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## CONCLUDING MATERIAL

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DOE-EH-52

**Project Number:**

SAFT-0055

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LANL

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1910-0900

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